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DESIGN OF ABSORPTION/DESORPTION TOWERS

MASS TRANSFER AND PRESSURE DROP DATA
FOR

PLASTIC PACKINGS

SVEN-GUNNAR TRULSSON

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AVHANDLING FÖR TEKNISK DOKTORSEXAMEN



**DEPARTMENT
OF
CHEMICAL ENGINEERING**

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Mass Transfer and Pressure Drop Data
for
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When designing packed towers mass transfer and pressure drop data are essential. Such data have been determined for the three plastic packings, Intalox saddles, Tellerettes, and Plasdek, operated countercurrently. Liquid-film mass transfer coefficients were determined by absorption and desorption of CO_2 in water, and gas-film mass transfer coefficients were determined by absorption of SO_2 in NaOH-solutions. The pressure drop and flooding points were measured for the flow of air and water. Finally, overall mass transfer coefficients were obtained by absorption and desorption of ammonia from water. By this operation ammonia can be recovered from waste water in connection with the ANAMET-process.

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Mass Transfer and Pressure Drop Data
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This thesis consists of this summary and of the following papers:

- I. Trulsson, S.G., Wimmerstedt, R.
"Liquid-film Mass Transfer Coefficients for Three Different
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- II. Trulsson, S.G.
"Gas-film Mass Transfer Coefficients for Three Different
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- III. Trulsson, S.G.
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- IV. Trulsson, S.G.
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List of Contents

	Page
1. Introduction	1.
2. The Packings	3.
3. Basic Design Concepts for Packed Towers	4.
4. Summary of the Present Work	6.
Part I - Liquid-side Coefficients	6.
Part II - Gas-side Coefficients	7.
Part III - Pressure Drop	8.
Part IV - Ammonia Desorption/Absorption	9.
5. Future Work	11.
Acknowledgements	11.

1. INTRODUCTION

When designing packed columns for use in air pollution control, or as an integrated part of a process, a knowledge of the performance of different packings is vital. After modern packings were introduced in 1915 by Raschig, a large number of different types have appeared on the market. The general idea with a packing is to generate as effective contact between the gas- and liquid phase as possible. This is achieved by distributing the liquid phase in a thin film and by promoting turbulence in the two phases, while still trying to keep the pressure drop at a low level.

The correct choice of packing for a specific installation is by no means a simple task. In figure 1 the most important points to be considered in the selection are given.

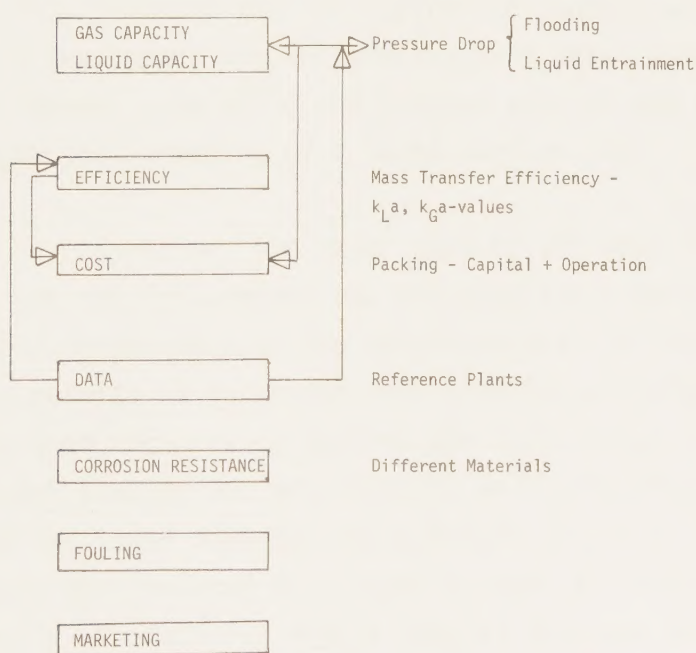


Figure 1. Criteria for selecting packings

Clearly no "best" packing can generally meet all these criteria. Some of the points, like capacity, efficiency, pressure drop, and total cost, are calculable provided that data for the specific packing are available. For many of the most effective packings there is, however, a lack of such data in the literature. Other points like marketing,

including know-how and technical assistance, are left to the subjective judgement of the design engineer.

In this thesis, performance data are given for three modern, 0.025 m nominal size, plastic packings: Intalox saddles, Tellerettes, and Plasdek. They were operated countercurrently in towers with a diameter of 0.3 m and at packing heights that could be varied up to 3 m.

2. THE PACKINGS

In air pollution control, where acidic or alkaline absorbents are often used, plastic seems to be a good construction material, due to its chemical resistance. Furthermore, often large gas quantities have to be handled. Both plastic Intalox saddles, Tellerettes, and Plasdek have high gas capacity, as is obvious from the manufacturers' pressure drop curves. They are also often very competitive as packing devices in many other gas transfer applications where the chemical system and temperature permit plastic to be used. However, the lack of efficiency data in the literature, especially for Tellerettes and Plasdek, have made them less used than expected.

In the first part of this thesis, the three packings are presented in Figure 1 and Table 1. The three packings have about the same geometrical properties, but the manner of operating is quite different. In a bed of Tellerettes the liquid is distributed as droplets, while a thin liquid film, or, rather, small rivulets, are formed in a bed of the others. This reflects the two theoretical models used when the packings were developed; the Tellerettes were based upon a high liquid surface renewal rate while the Intalox saddles and the Plasdek are based upon the formation of a large surface area.

In this investigation, the major part of the data for the Intalox saddles and the Tellerettes was obtained for a more densely packed tower than recommended by the manufacturers. (I, Table II). This probably results in increased mass transfer efficiency, but also pressure drop and cost of packing per cubic meter will be higher. The Plasdek packing was manufactured in blocks with a diameter equal to the tower diameter and a height of 0.3 m. When necessary, the blocks were reduced to a lower height, as desired. The Plasdek is normally manufactured with a special drainage tip system at the bottom of each block. The packing examined here had no such drainage tips.

3. BASIC DESIGN CONCEPTS FOR PACKED TOWERS

The design of a packed tower can, generally, be divided into three steps:

- Determining the tower diameter
- Establishing the tower height
- Selection of tower internals

The basis for determining the tower diameter is the pressure drop of the selected packing. When the flow of gas and liquid is countercurrent, the gas handling capacity is limited by the flooding velocity. The flooding velocity is determined from pressure drop measurements and can generally be estimated from a flooding point correlation. The best operating condition for the tower is at the loading point, which is established by a suitable reduction of the flooding velocity. From the flow rates at loading, the tower diameter is determined. In section III of this thesis a more comprehensive study of pressure drop and flooding for the three packings is described.

The basic concepts involved in establishing the tower height are the driving force and the mass transfer coefficient for the transfer operation. Each system has its own mass transfer coefficient and driving force. The mass transfer coefficient varies with the choice of packing; a packing with higher turbulence-promoting capacity and larger effective interfacial area has a higher efficiency. From pilot-plant experiments, the transfer coefficient can be determined for any particular gas absorption system. From these experiments a full-scale tower can be designed. In part IV of this thesis, such experiments are presented for ammonia desorption from water. This operation is of particular interest in connection with the ANAMET process, for the recovery of nitrogen from protein containing waste water.

With the two-film theory, and the models for gas absorption accompanied by chemical reactions, an opportunity was given to estimate overall mass transfer coefficients. According to the film theory, the total resistance to mass transfer is confined to thin regions adjacent to the liquid-gas interface. This means that an overall mass transfer coefficient can be derived from one gas-film coefficient, $k_G a$, and one liquid-film

coefficient, $k_L a$, together with an enhancement factor, E , due to a reaction, and the physical solubility of the gas in the absorbent, H , as

$$\frac{1}{K_G a} = \frac{1}{k_G a} + \frac{1}{H \cdot E \cdot k_L a} = \frac{1}{H \cdot K_L a} \quad (1)$$

where $K_G a$ and $K_L a$ are the overall mass transfer coefficients.

Clearly, the overall coefficient can only be obtained from equation (1) if the order and rate of the gas-liquid reaction are known. For some commercially important absorption systems this is so. Furthermore, since E varies with composition along the tower, $K_G a$ (or $K_L a$) will generally also vary significantly. This must be taken into consideration when the tower height is determined.

The individual mass transfer coefficients can be regarded as packing characteristics. They can be determined as volumetric coefficients by experiments with a system that is fully gas-film controlled and with a system that is fully liquid-film controlled. For a few packings like Raschig rings and Berl saddles, they can also be obtained as true mass transfer coefficients from empirical modifications of relationships developed from the analogies between heat and mass transfer. For the tower height determination, the resulting overall mass transfer coefficient must be multiplied with the effective transfer area of the packing, a_{eff} . By chemical absorption methods, a_{eff} can be determined. However, the most reliable method seems to be to directly establish the volumetric coefficients as is done for the three packings in section I and II of this thesis. These $k_G a$ - and $k_L a$ -values are valid only for the systems tested. They vary with physical properties, such as diffusivity and viscosity. However, by using a suitable mass transfer model, which takes account of these variations, the coefficients can be recalculated for other systems.

Selection of tower internals is mostly a matter of experience. Manufacturers generally offer guidelines for the selection of gas and liquid distributors, wall-wipers and liquid redistributors, and on packing supports. Often it is also necessary to install a mist eliminator at the top of the tower. Section I contains some guidelines concerning the need of redistributors. It is also concluded that it is essential to have a good liquid distributor when Tellerettes are used.

4. SUMMARY OF PRESENT WORK

Part I: Liquid-side coefficients

Liquid-film mass transfer coefficients were obtained by absorption and desorption of CO_2 in deionized water. Since carbon dioxide is sparingly soluble in water, this system is regarded as fully liquid-film controlled. The reaction of CO_2 and water to HCO_3^- and CO_3^{2-} had no influence on the absorption rate in deionized water. This was tested by the criteria for slow and very slow reactions. By developing a special model, which took account of the equilibrium carbon dioxide concentrations as the pH changed along the tower, a $k_L a$ -value was obtained that included the effect of hydration. This $k_L a$ -value was equal to the $k_L a$ -value obtained from equations for physical absorption.

In the very wide range of operating conditions tested, about the same $k_L a$ -values were obtained for the three packings. For absorption purposes the $k_L a$ -values were somewhat higher for the Intalox saddles, probably because of the densely packed bed.

The $k_L a$ -values varied between 0.01 and 0.05 s^{-1} , depending mainly on liquid load. At lower liquid loads the dependency was more pronounced than at higher loads, because the increase in effective area diminishes. In spite of the full liquid-film control a gas-flow dependency was also noticed. It was concluded that this was the result of back-mixing of the gas phase when the upward gas velocity was much smaller than the downward liquid-film velocity. Provided the gas velocity is sufficiently high, the $k_L a$ -values are independent of the gas velocity.

Lower $k_L a$ -values were obtained at greater packing heights. This was probably due to back-mixing of the gas phase, although maldistribution of the liquid could also have an influence. At lower packing heights, $k_L a$ -values obtained for desorption were higher than those obtained for absorption. This could be an effect of the hydration reaction. At greater packing heights, both absorption and desorption exhibited the same $k_L a$ -values. A plausible explanation of this is that the effect of back-mixing is more severe in desorption operations.

From a literature survey, $k_L a$ -values obtained for packings of the same nominal size seem to be of the same order of magnitude. Quantities such as temperature, packing height, and packing density may each have an influence on the liquid-side mass transfer efficiency comparable to the influence of the particular packing type.

Part II: Gas-side coefficients

Gas-film mass transfer coefficients were obtained by absorption of SO_2 in caustic soda solutions. With a low SO_2 -concentration in the gas phase and high pH, this system fulfils the criterion for full gas-film control. A wide range of gas and liquid loads was tested. The gas load varied from 0.6 to 3.3 $\text{kg/m}^2 \cdot \text{s}$ and the liquid load from 2 to 50 $\text{kg/m}^2 \cdot \text{s}$. These wide ranges make the result especially valuable in designing air pollution control equipment.

The resulting $k_G a$ -values vary between 0.7 and 4 $\text{kmole/m}^3 \cdot \text{s MPa}$ depending on gas and liquid flow rates and type of packing. The Tellerettes have the superior mass transfer efficiency; up to 50 percent better than the Intalox saddles at low gas flow rates. At higher flow rates the difference in $k_G a$ -values of the three packings becomes smaller, because of different gas and liquid flow rate dependency. At comparable flow rates, the Plasdek has the lowest $k_G a$ -value, while at the loading point, which is at higher flow rates for the Plasdek, the Plasdek has the highest efficiency.

A rather sharp transition was noted at a minimal wetting rate. Most literature data are established at or below this minimal wetting rate. Below the minimum wetting rate, the available transfer area is badly used. When extrapolating such data to higher liquid loads, care must be taken not to underestimate the tower height.

The mass transfer efficiency increases markedly with the gas load for gas-film controlled systems. It is thus preferable to use a high gas load. Since the residence time of the gas diminishes at higher gas loads, a higher tower will be necessary. The total packing volume will, however, be smaller. It is thus a matter of balancing increased pressure drop, as the gas velocity is getting closer to the flooding velocity, against decreased tower and packing costs. For the Plasdek, with its high flooding velocity, higher gas loads should preferably be used.

Published $k_G a$ -values have been obtained with a variety of systems. For gas absorption in packed columns there are strong indications that the true gas-film coefficients for different systems are proportional to $\sqrt{D}$. When data obtained in this investigation are compared to literature data, adjusted with $\sqrt{D}$, some difference is still noted. Most likely this is due to end-effects not accounted for when establishing the $k_G a$ -values.

Part III: Pressure Drop

Pressure drops for countercurrent flow of air and water as well as liquid holdup were measured for the three packings. The result shows that the Plasdek has the superior capacity and the lowest liquid holdup. The Intalox saddles have the highest liquid holdup but the lowest capacity.

The wet pressure drops were correlated with a model based upon the interaction of liquid holdup on dry pressure drop. The liquid holdup was correlated with a film flow model and the dry pressure drop with a slightly revised Ergun type of equation. Only pressure drops at working conditions below loading and at moderate liquid loads could be correlated in this way. This is mainly due to the influence of gas load on liquid holdup above loading. From results presented in the literature it was anticipated that the correlation could be used to predict pressure drops at the same working conditions for more relevant mass transfer systems, i.e. for other gas and liquid combinations as well as for the packings at other sizes. However, if pressure drop curves are available for the particular system they should preferably be used.

The generalized pressure drop correlation, as recently reworked by the Hydronyl Company, could not be used to correlate the present data. At certain flow conditions the deviation could very well be above one hundred percent. The packing factor, F , as determined from the measured pressure drops, varied considerably with flow rate and pressure drop level.

The flooding points for the Intalox saddles and the Tellerettes could be correlated reasonably well using methods proposed in the literature.

However, none of them could be used to correlate the flooding points for the Plasdek. With its very open structure, this packing seems to have hydrodynamic properties that largely resemble stacked packings'. At low liquid loads the flooding pressure drop is also very low, because flooding virtually only occurs at the contact zones between the packing blocks. These properties can be accounted for by the introduction of a "flooding" factor. However, even if the correlations are rather inaccurate in determining the exact flooding velocity, the use of a sufficiently large reduction factor to obtain the loading velocity results in a safe operating condition. The pressure drop at this loading condition must then generally be determined from experiments. Alternatively, it may be estimated from a pressure drop correlation, however with somewhat lower accuracy.

Part IV: Ammonia Desorption/Absorption

Ammonia can be removed from waste water in the ANAMET-process by counter-current desorption with air in a packed column. By subsequent absorption of ammonia in an acid, many problems of direct air stripping are avoided and a valuable fertilizer is produced.

Since ammonia is very soluble in water, a packing that permits high gas load must be used. Even then the liquid load must be low, preferably below the minimum wetting rate. Three packings were tested: Plasdek, Tellerettes, and Sulzer BX. Sulzer BX is a packing with the same configuration as the Plasdek. It has twice as large a geometric area (see table 1) and has very good wetting properties at low liquid loads because of a special woven packing surface.

The desorption design data, as obtained for the three packings, were very low. A reason for this is the extremely low liquid load used. The wetting of the packing may therefore be bad. For ammonia desorption it is thus essential to establish the mass transfer efficiency through pilot-plant experiments.

The absorption took place in phosphoric acid at a low pH. Essentially gas-film coefficients were thus obtained. When compared to $k_G a$ -values as obtained when absorbing SO_2 , the ammonia data was higher. After corrections for end-effects and diffusivity the difference was about 15 per-

cent. The influence of pH and ionic strength were studied. At higher pH, the mass transfer coefficient diminished, due to an increased liquid-film resistance. At increased ionic strength higher mass transfer coefficients were obtained, probably because of altered reaction conditions and increased mass transfer area.

From an economic analysis it was concluded that a method of desorption and absorption of ammonia in packed columns was comparable to other ammonia reduction methods, like biological denitrification. It is also interesting to note that the cost of towers and packing is only a small part of the total cost.

5. FUTURE WORK

The design of mass transfer equipment for absorption/desorption purposes is a complex problem. The present data have been used to design commercial plants successfully. However, packings of larger nominal sizes generally have to be used. The determination of performance data for such packings is planned. For this purpose a new pilot-plant with a larger diameter is being built in the department. Investigations are also planned concerning the influence of packing density on the performance, and concerning the severe back-mixing in a bed of Plasdek at higher liquid loads. Furthermore, a theoretical explanation of the difference in mass transfer efficiency obtained for absorption and desorption, especially in the case of transferring ammonia remains still to be put forward.

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LIQUID-FILM MASS TRANSFER COEFFICIENTS
FOR THREE DIFFERENT TYPES OF PLASTIC
PACKINGS

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Liquid-Film Mass Transfer Coefficients for three Different Types of Plastic Packings.

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Liquid-Film Mass Transfer Coefficients have been determined for three types of plastic packings: Intalox saddles, Tellerettes and Plasdek. This was accomplished by absorbing and desorbing carbon dioxide in water. A wide range of operating conditions was examined; the liquid range probably covers most industrial applications, while the gas rate varies from below loading condition to flooding condition.

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ABSTRACT

Liquid-film mass transfer coefficients have been determined for three different plastic packings: plastic Intalox saddles, Tellerettes and Plasdek. This was accomplished by absorbing and desorbing carbon dioxide in deionized water. A wide range of operating conditions was examined; the liquid range probably covers most industrial applications while the gas rate varies from below loading condition to flooding condition.

In spite of the fact that the three packings have quite different geometrical layouts and modes of operation, they exhibit roughly the same liquid side mass transfer coefficient. A survey of existing literature shows that temperature, column height and diameter may have a greater influence on the mass transfer efficiency than the particular packing type.

The liquid flow rate showed a marked effect on the mass transfer coefficient, larger coefficients being obtained at higher liquid rates. Gas velocity also was observed to have an influence when downward liquid velocity was much greater than upward gas velocity. When this was so, back-mixing of the gas was believed to occur. The packings showed different tendencies to back-mix the gas. Consequently the mass transfer efficiency of the Tellerettes and the Plasdek was more affected by back-mixing than that of the Intalox saddles.

The present data make it possible to study the effects of direction of mass transfer, i.e., absorption versus desorption, and of packing height. In industrial practice, where towers greater than 1 meter are often used, the effect of direction may be disregarded. The height effect, on the other hand, necessitates the installation of redistributors. The intervals between redistributors found in this investigation varied from 2-7 times the column diameter, depending both on packing type and on process.

Table of Contents

	page
1. Introduction	1.
2. Methods of Investigating Liquid-Side Packing Efficiency	3.
3. Description of the CO ₂ /Air-Water system	5.
4. Experimental Equipment	7.
5. Analyses	10.
5.1 Liquid Side Analyses	10.
5.2 Gas Side Analyses	12.
6. Method of Investigation	15.
6.1 Packing Installation	15.
6.2 Liquid and Gas Rates	15.
6.3 Experimental Procedure	16.
7. Evaluation of Data	18.
7.1 The Mass Balance	18.
7.2 Calculation of the Mass Transfer Coefficient	19.
7.2.1 Ignoring the Hydration	20.
7.2.2 Considering the Hydration	20.
7.3 Accuracy in Determined Mass Transfer Coefficients	22.
8. Results	25.
8.1 Effects of Hydration	25.
8.2 Effects of Liquid and Gas Rates	29.
8.3 Comparison of the three Investigated Packings	31.
8.4 Comparison of Other Packings	32.
8.5 Comparison of Transfer Data Obtained by Absorption and by Desorption	33.
8.6 Effects of Packing Height on the $k_L a$ -values. Wall Flow	34.
9. Conclusions	49.
Acknowledgements	51.
Nomenclature	52.
References	54.

Figures

No.	Title	Page
1.	Intalox Saddles, Tellerettes, Plasdek	2.
2.	pC-pH diagram of $\text{CO}_2\text{-H}_2\text{O}$	6.
3.	Experimental Equipment	9.
4.	Liquid distributor	9.
5.	Gas distributor	9.
6.	Cross section of the pCO_2 -electrode	13.
7.	pCO_2 -pH calibration diagram	13.
8.	Calibration cell	13.
9.	The liquid analyzer cell	14.
10.	Infrared gas analyzer	14.
11.	Gas calibration diagram	14.
12.	The tower divided into incremental sections	23.
13.	Operating and equilibrium lines	24.
14-15	k_La -values obtained with Intalox Saddles	39.
16-18	k_La -values obtained with Tellerettes	40.
19-21	k_La -values obtained with Plasdek	41.
22-23	Comparison of k_La -values from the three packings	42.
24.	Comparison with results of Sahay and Sharma (ref. 10)	42.
25.	Literature data, Raschig rings	43.
26.	Literature data, Raschig rings corrected to 25 C	43.
27.	Literature data, Intalox Saddles	44.
28.	Literature data, miscellaneous packings	44.
29.	Effect of process, Intalox saddles at $z = 0.75$ m	45.
30.	Effect of process, Tellerettes at $z = 0.75$ m	45.
31.	Effect of process, Plasdek at $z = 0.75$ m	45.
32.	Effect of process, Plasdek at $z = 1.5$ m	46.
33.	Effect of process, Plasdek at $z = 3$ m	46.
34.	Effect of packing height, Intalox Saddles	47.
35.	Effect of packing height, Tellerettes	47.
36.	Effect of packing height, Plasdek	47.

Tables

No.	Title	Page
1.	Geometrical data for the packings	2.
2.	Number of packing pieces per cubic meter of packing	17.
3.	Maximum error in $k_L a$	23.
4.	Standard deviation of $k_L a$	24.
5.	Tests of very slow reaction	38.
6.	Tests of slow reaction	38.
7.	γ in $k_L a = \alpha \cdot (L)^\gamma$; $L < 15 \text{ kg/s} \cdot \text{m}^2$	48.
8.	Liquid film data reported in literature	48.
9.	Wall flow	48.

1. INTRODUCTION

The packed column is a widely used piece of chemical equipment. When designing a tower, a great many types of packings may be considered. However, the selection is limited because reliable and general design data are reported in the literature for only a few types of packings.

The efficiency of a packing is primarily determined by turbulence promoting capacity, both in liquid film and gas core, effective specific area, wetting, and distribution properties. The efficiency can be expressed by the $k_L a$ - and $k_G a$ - values. These values are easily determined by making measurements on a physical mass transfer system that is fully liquid-film controlled and on a system that is fully gas-film controlled.

In this preliminary report liquid film mass transfer data ($k_L a$ -values) for three packings are presented. The packings are plastic Intalox saddles, Tellerettes, and Plasdek. These packings are all made of non wetting plastics, i.e. they all exhibit about the same wettability. Their distribution properties, however, are quite different, as will be demonstrated. The three packings are rather newly developed, and for two of them there exist almost no data in the literature. In figure 1 the packings are shown and in table 1 some geometrical data from the manufacturers are displayed.

In a succeeding paper gas film mass transfer data ($k_G a$ -values) for these packings will be reported.

Table 1.

Geometrical data for the packings			
Type	nominal size mm	a $\frac{m^2}{m^3}$	ϵ %
Plastic Intalox saddles	25	207	91
Tellerettes	25	180	87
Plasdek 12060	--	225	97

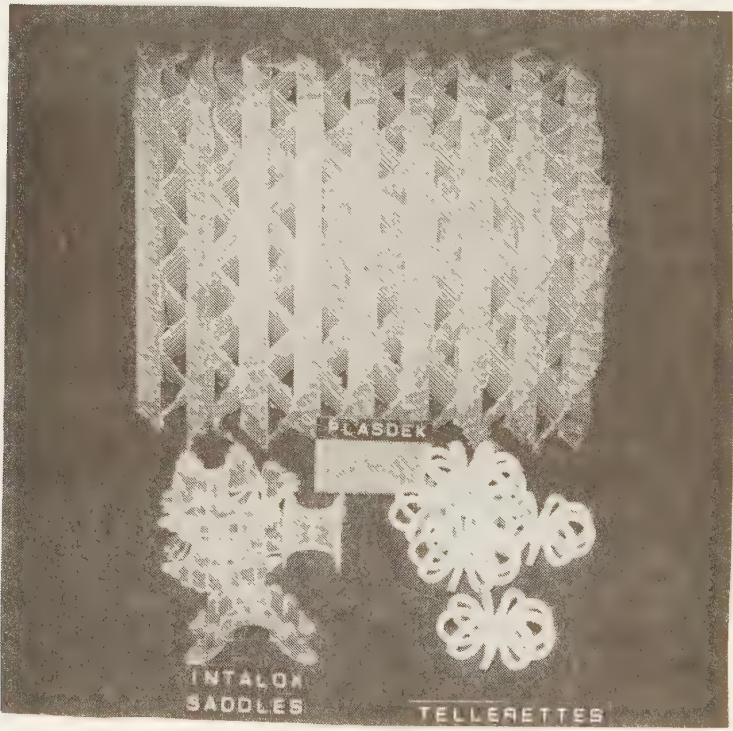


Figure 1.

Plastic Intalox Saddles, Tellerettes, Plasdek

2. METHODS OF INVESTIGATING LIQUID SIDE PACKING EFFICIENCY

A method often used to establish the efficiency of a packing is to make mass transfer studies on a system with a chemical reaction. Manuals showing the efficiency of different packings generally use the $\text{CO}_2/\text{air-NaOH}$ ¹ system. Another system used is CO_2 absorption in solutions of various ethanolamines. Teller, ² for instance, has used this system to examine the Tellerettes. These systems have direct industrial applications, e.g. gas cleaning in manufacturing ammonia or hydrogen.

The mass transfer efficiency so obtained may be recalculated by the enhancement factor in order to give a physical mass transfer coefficient, a procedure which is outlined for instance by Yoshida ³. However two major problems exist. One is that the systems are not entirely liquid-film controlled which therefore makes it necessary to estimate the gas-film resistance. The other problem is that system properties of the actual ionic solution, such as solubility, diffusivity, and rate-constants for the reaction often must be estimated by standard methods ⁴.

A more straight-forward method is to determine the $k_L a$ -value directly by using a pure physical mass transfer system. The comparison between packings may then be made directly. If desired the $k_L a$ -value may, in general, be more accurately recalculated to estimate the mass transfer of another gas-liquid system.

Commonly used physical absorption-desorption systems are $\text{O}_2\text{-H}_2\text{O}$ ^{5,6,7,11}, $\text{H}_2\text{-H}_2\text{O}$ ¹¹ and $\text{CO}_2\text{-H}_2\text{O}$ ^{3,8,9,10,11,12,13,14,15,16}. A summary of these investigations on packings with 25 mm nominal size can be found in table 8.

Oxygen, hydrogen, and carbon dioxide are all slightly water soluble. If the gas is slightly soluble in the liquid then the mass transfer is generally liquid-film controlled. However, there is an increasing gas-film resistance when the concentration of the absorbable component in the gas-phase decreases. For instance, when absorbing a 10 % CO_2 -air mixture in water, there will be about 2 % gas-film resistance.

The difficulty with pure physical mass transfer systems is that the liquid will be saturated with the soluble gas component if the packing depth is too great. Since rather small packing depths must be used the degree of conversion will be low and the need for accurate analyses is essential.

In this study the $\text{CO}_2/\text{air-H}_2\text{O}$ system is employed. This system is almost entirely liquid film controlled. Data are available for some packings in the literature and accurate analyses can be performed.

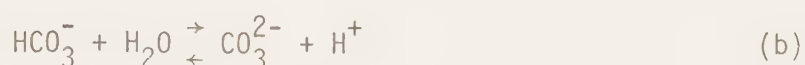
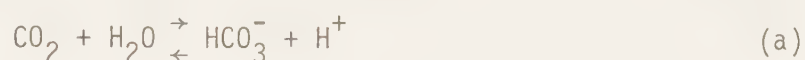
3. DESCRIPTION OF THE CO₂/AIR-WATER SYSTEM

Carbon dioxide is slightly soluble in water. The Henry's law constant of carbon dioxide in pure water at about atmospheric pressure is given by Danckwerts and Sharma¹⁷ as:

$$\log H_e = \frac{1140}{T} - 4.30 \quad (1)$$

(T in K). This expression is in agreement with data given by Perry¹⁸.

Carbon dioxide reacts with water in a two step equilibrium reaction



(The amount H₂CO₃ formed is negligible¹⁷.)

The stoichiometric ionization constants for (a) and (b),

$$K_1 = \frac{|\text{H}^+| \cdot |\text{HCO}_3^-|}{|\text{CO}_2|} ; \quad K_2 = \frac{|\text{H}^+| \cdot |\text{CO}_3^{2-}|}{|\text{HCO}_3^-|} \quad (2)$$

have the following value at 298 K.

$$K_1 = 4.437 \cdot 10^{-7} ; \quad K_2 = 4.67 \cdot 10^{-11}$$

Danckwerts and Sharma¹⁷ also give expressions for the temperature dependence on K₁ and K₂:

$$\log K_1 = - 3404.7/T + 14.843 - 0.03279 \cdot T \quad (3)$$

$$\log K_2 = - 2902.4/T + 6.498 - 0.0238 \cdot T \quad (4)$$

In figure 2 the logarithms of the concentrations of the various protolytic species are plotted as a function of $-\log|\text{H}^+|$. It is obvious from this figure that the bicarbonate content rapidly decreases with decreasing pH. Tap water with a pH of about 7 has a higher bicarbonate content than the deionised water used in this investigation, which had a pH of about 6.

The reaction (a) is a pseudo-first-order reaction in the forward direction. It is a slow reaction. The rate constant is given by Danckwerts and Sharma¹⁷ as:

$$\log k_{H_2O} = 329.850 - 110.541 \cdot \log T - 17265.4/T \quad (5)$$

At 298 K k_{H_2O} has a value of 0.026 s^{-1} . The reverse reaction of (a) is a second order reaction. The rate constant is given by (at 298 K)

$$k_{H_2O}^{-1} = \frac{k_{H_2O}}{K_1} = 58600 \quad (6)$$

At equilibrium the forward rate of the reaction is equal to the reverse rate.

The effect of the hydration reaction is to reduce the back-pressure of carbon dioxide in the absorbing liquid i.e. the absorption is not a purely physical one. In this investigation the fact that pure water (deionized) was used diminished the effect of hydration. The reaction has never-the-less been taken into account when calculating the k_L -values. Subsequently the hypothesis will be tested that the reaction is too slow to influence the absorption-desorption process.

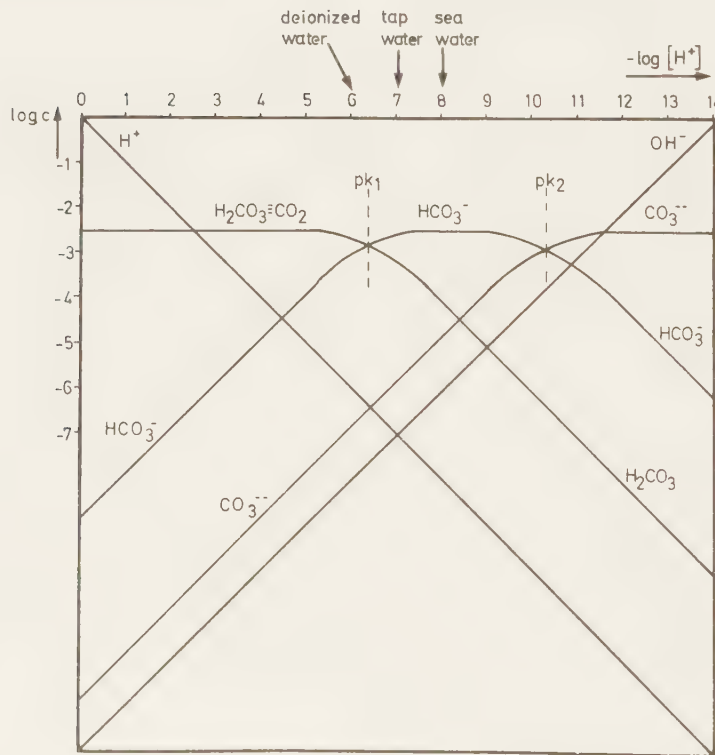


Figure 2. pC-pH diagram of $\text{CO}_2\text{-H}_2\text{O}$.

4. EXPERIMENTAL EQUIPMENT

The experimental equipment consisted mainly of two towers, one for absorption and one for desorption. Thus the liquid could be recirculated. This was a necessity since deionized water was available only in limited quantities. Figure 3 schematically shows the towers with the gas and liquid streams. The design permitted packings to be changed rather easily by lifting the towers with a travelling crane.

The towers had an inside diameter of 0.3 m and were 3 m high. Below each of the towers a liquid storage tank was located. From this tank the liquid was pumped by a standard pump to the top of the second tower.

The air to the desorber was taken directly from the room and blown by a fan to the bottom of the desorption tower. The gas entrance to the absorber was situated adjacent to the exhaust of the desorber. As a consequence the gas blown to the absorber contained about the same amount of carbon dioxide as did the exhaust gas from the desorber. The gas was made up to desired inlet concentration with pure carbon dioxide from a gas cylinder. To ensure proper mixing of the gas a mixing length of about 3 m was provided.

The towers were constructed of polyester. This material had the advantage of being both corrosion- and impact-resistant. Furthermore polyester, unlike glass, is a non-wetting material. The amount of liquid flowing down the wall thus will be smaller in a polyester tower than in a glass tower. The main disadvantage of polyester towers was that the flow pattern in the towers could not be visualized. The insertion of three small windows of plexiglass, however, made it possible to inspect the flow pattern.

The flow-measurements were made by calibrated orifices. The liquid streams were individually measured by two orifices, one for the absorber and one for the desorber. The gas flow was measured by interchangeable orifices. Thus the pressure drop was kept at an acceptable level while still yielding good readings. All orifices fulfilled the requirements of the Swedish orifice standard.

The liquid distributor used was designed to operate at both low and high irrigation rates. Figure 4 shows the liquid distributor. The number of ho-

les (21), is in rather good agreement with the rule-of-thumb given by Eckert¹⁹. Between the wall of the tower and the outer hole a passage for the gas was left. This probably did not introduce any severe maldistribution of the liquid, since the liquid splashed on the top of the packing and there is some selfdistribution in the packing. The distributor was always placed immediately above the packing.

In figure 5 the gas distributor is shown. This special design permitted the gas to enter the whole crossarea very evenly. The water passed down via the annular space between the outer wall and the gas pipe, and into the liquid seal, without any leakage into the gas pipe.

A check of the liquid distribution and the selfdistribution in the packing was made by measuring the wall flow. This was done at the bottom of the desorber by collecting the water flowing through an area adjacent to the wall. This "wall area", made up of an annulus slot section, consisted of 10 % of the total "tower area". Special holes were drilled, from which the wall flow was collected and measured.

The packing support used for plastic Intalox saddles and Tellerettes was of bar-grid type. It was designed to carry ceramic packings. The support had a free area of about 70 %. The packing support could be removed, and in fact no support was used during the runs with the Plasdek.

The temperatures of the gas and liquid streams were measured by thermocouples. The temperature of the liquid was measured in the storage tanks. The wet and dry bulb temperatures of entering and leaving gas were also measured. The humidity of the leaving gas was always in equilibrium with the liquid.

The liquid temperature could be adjusted in two ways. To raise the temperature before starting a test, immersion heaters were used in the storage tanks. To keep it constant during a test cold water was let into the tanks. The liquid temperature was adjusted to 298 K.

The sampling system was designed so that end-effects were minimized. Liquid samples were taken on entering and leaving liquid. The entering liquid sample was taken from the pipe to the tower. Immediately below the support plate the leaving liquid sample was taken via two glass funnels. The samples were led by airtight flexible tubes (PVC) to the liquid analyzer cell. In this

way the samples nowhere came into contact with the surrounding air before the analyses.

Gas samples from the desorber were taken only on leaving gas. The inlet concentration was known to be that of normal atmosphere. From the absorber samples were taken on both incoming and outgoing gas. The lower sampling point was placed immediately below the gas distributor. The upper sampling point could easily be adjusted so that it was always placed directly above the packing, independent of the packing height. Only one point of the cross-section was examined. All gas samples were dried by silica gel in a glass tube at the end of the hose. The dried gas samples were then led by air-tight flexible tubes to the gas analyzer.

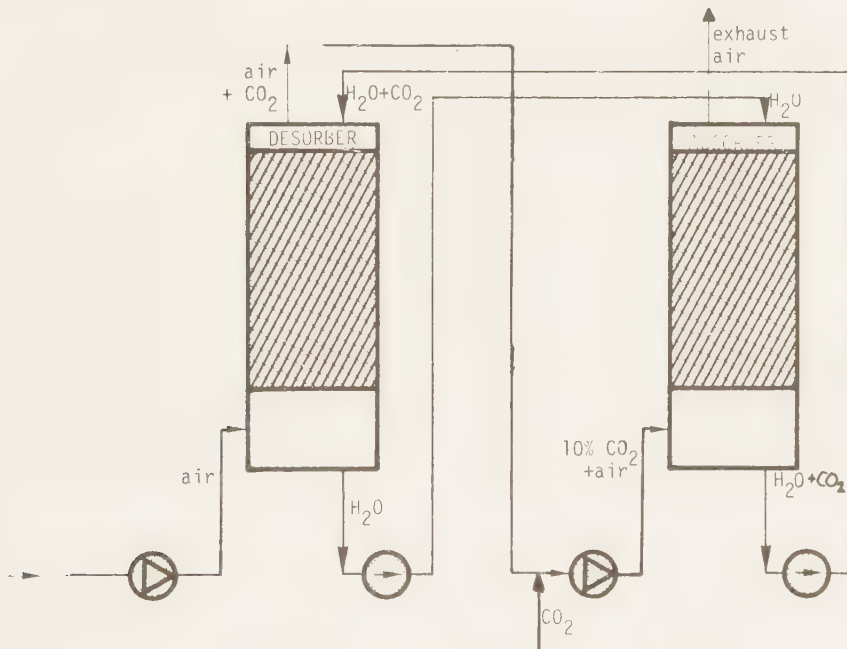


Figure 3. Experimental Equipment.

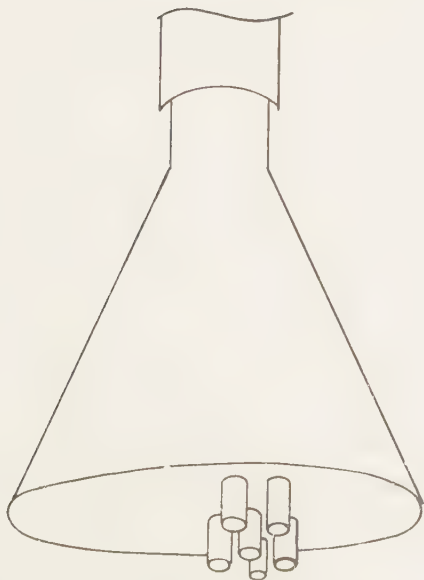


Figure 4. Liquid distributor.

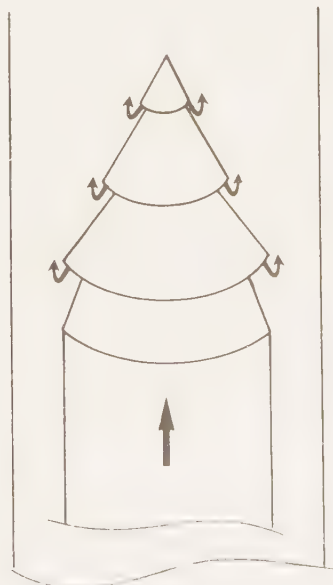


Figure 5. Gas distributor.

5. ANALYSES

Both liquid and gas streams were analysed. This was done in order to check the mass balance. Both analyses were continuous and did not involve titration, which is commonly used in absorption-desorption studies of CO_2 in H_2O .

5.1 LIQUID SIDE ANALYSES

According to Park²⁰ the total composition of a carbon dioxide - pure water mixture is known if for instance pH and partial pressure of carbon dioxide, p_{CO_2} , are measured. pH is accurately measured by a glass electrode. The p_{CO_2} can be measured by an ion-selective electrode. The electrode used was a Radiometer E5037 electrode. It was originally developed to measure carbon dioxide content in human blood.

The electrode essentially consists of a glass electrode, (an arched McInnes type), combined with a silver/silver chloride reference electrode. These electrodes are mounted in a special jacket which is filled with a solution of NaHCO_3 and NaCl . At the bottom of the jacket a membrane, either of teflon or of silicon rubber, is fitted. The latter material will give faster response and was used in this investigation. In figure 6 a longitudinal section of the p_{CO_2} electrode is given.

The operating characteristics of the electrode have been described by several authors^{21,22}. The principle is that molecular carbon dioxide, for instance dissolved in water, diffuses through the membrane. The pH of the jacket solution will change depending on the carbon dioxide content

$$K_1 = \frac{|\text{HCO}_3^-| \cdot |\text{H}^+|}{|\text{CO}_2|} \quad (2a)$$

The partial pressure of carbon dioxide, which is in equilibrium with a water solution, is known from Henry's law

$$|\text{CO}_2| = \text{He} \cdot p_{\text{CO}_2} \quad (7)$$

The combination of these two equations will give:

$$\log (p_{\text{CO}_2}) = - \text{pH} + \log |\text{HCO}_3^-| + \text{p}K_1 - \log (\text{He}) \quad (8)$$

i.e., a linear dependence between the pH of the solution in the jacket and $\log (p_{\text{CO}_2})$ of the water solution exists, when equilibrium is established.

The signal from the p_{CO_2} electrode was in this investigation registered by an ordinary pH-meter. The pH-reading was converted to p_{CO_2} through use of a calibration chart. Figure 7 shows such a diagram. The calibration was done in a special cell with a small dead volume (fig. 8). Tempered gas of known concentration was introduced into this cell, which was maintained at 298 K. The small volume gave the desired gas concentration before the membrane was too dry. It appeared to be essential that the membrane remain wet during the calibration. Three different gases were used; 0.91 vol-%, 3.15 vol-%, and 7.55 vol-% carbon dioxide in nitrogen. The volumetric content was recalculated to p_{CO_2} by the equation

$$p_{\text{CO}_2} = \frac{(P - p_{\text{H}_2\text{O}}) \cdot \text{Vol}_{\text{CO}_2}}{100} \quad (9)$$

where P is the barometric pressure in MPa.

The liquid analyses were made in a glass cell, maintained at a constant temperature as shown in figure 9. To this cell liquid from the sampling points was slowly added. It is very important that the liquid sample does not come into contact with the surrounding air, and that no gas bubbles are entrapped. If this happens CO_2 immediately strips off and the analysis will be incorrect.

After liquid had flown through the cell some minutes pH, p_{CO_2} and temperature were measured. Due to the strong effect of temperature in the Henry's law constant, it was deemed necessary to take temperature measurements with a thermometer graded in 0.1 C.

The particular method of analysing the liquid allows pH to be measured with a precision of ± 0.01 , and p_{CO_2} with a precision of $\pm 2 \text{ ppm}^{20}$. The reproducibility of pH measurements was also very good. The p_{CO_2} measurements however did show somewhat poorer reproducibility. This was probably due to the calibration procedure.

5.2 GAS SIDE ANALYSES

In most previous investigations of carbon dioxide systems the gas has been analysed by an Orsat analyzer. This is a very tedious analytical method, and it does not allow continuous measurements to be made. In this investigation a continuous infrared analyzer was used (figure 10). It is well known that most gases exhibit characteristic light-absorption spectra. The analyzer used, MSA Lira 300, was designed with windows, detector, light-source, and filtering cell, so that the unique light-absorption band of carbon dioxide was measured. The cell length was designed for a calibration range of 1 % CO₂. In this range the Lambert-Beer's law of light-absorption was valid. This law was not valid for the concentration range used in this investigation (0-10 %). By altering the gain this range could be analyzed. However a calibration was then required, which resulted in a diagram like that of figure 11. The gases used were the same as those used for the p_{CO₂}-electrode calibration. In order to obtain accurate readings the amplifier signal was recorded. This also made it easier to identify the equilibrium of the mass-transfer process.

Dry gas from three different points of sampling were sucked individually into the analyzer. A gas sample was said to be analysed when the recorder showed a stable value. One analysis took about 5 minutes. This rather long time expenditure was mostly due to the dead volume of the tubes in the sampling system.

According to the manual of the instrument a maximum error of the magnitude of ± 1 percent of full scale (i.e. in this investigation ± 0.1 vol-% in each reading) may exist when a recording apparatus is used. Calibration tests performed each test day showed that the reproducibility was very satisfactory.

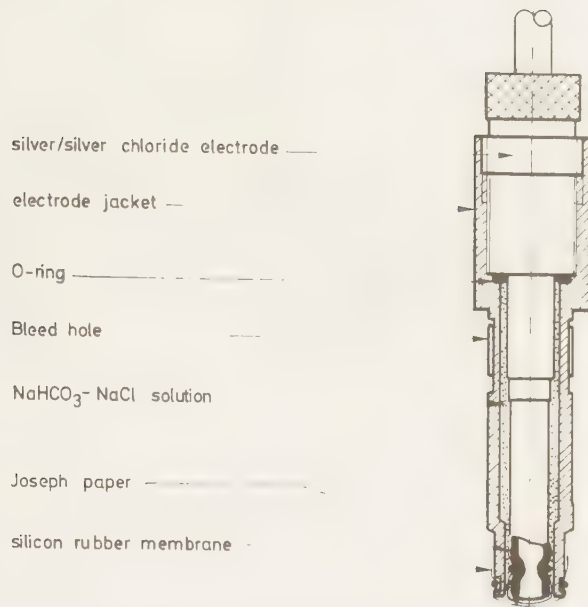


Figure 6. Cross section of the pCO_2 -electrode

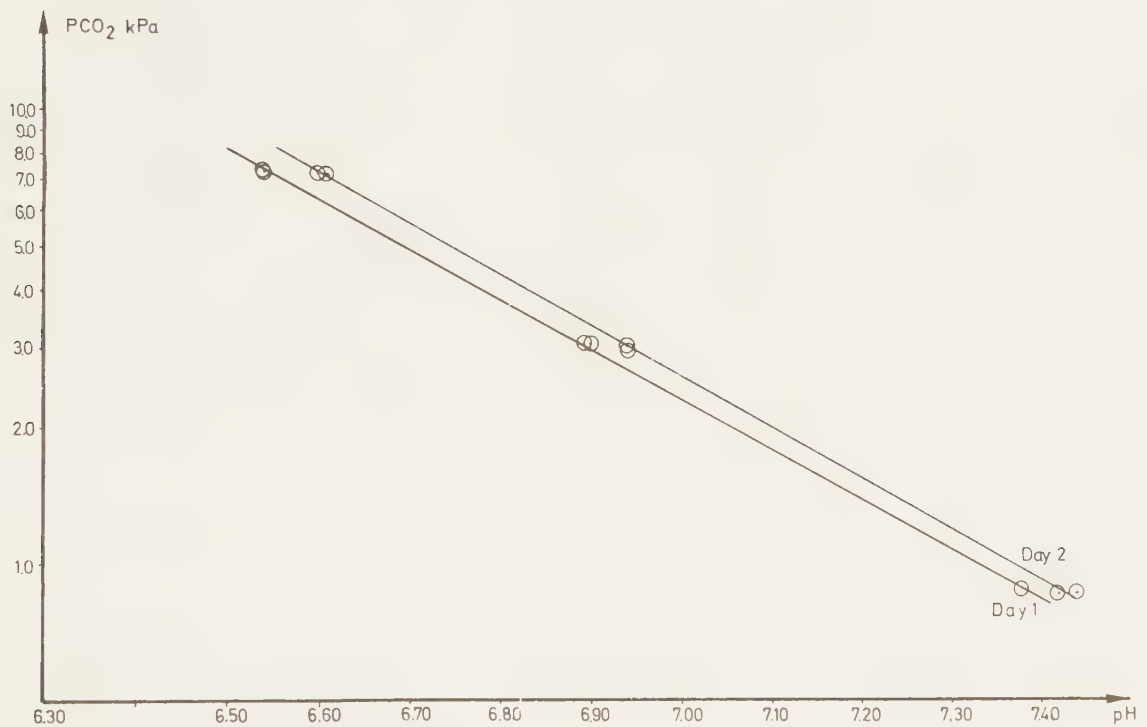


Figure 7. pCO_2 -pH calibration diagram.

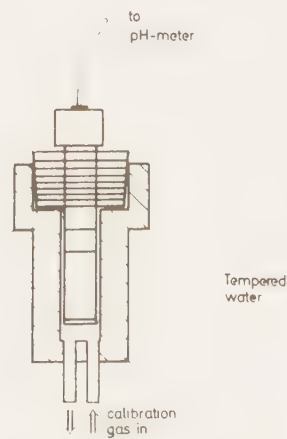


Figure 8. Calibration cell

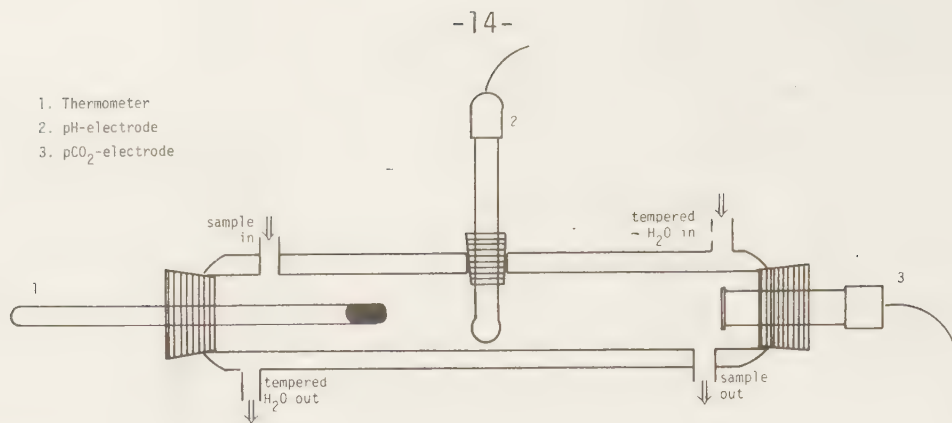


Figure 9. The liquid analyzer cell.

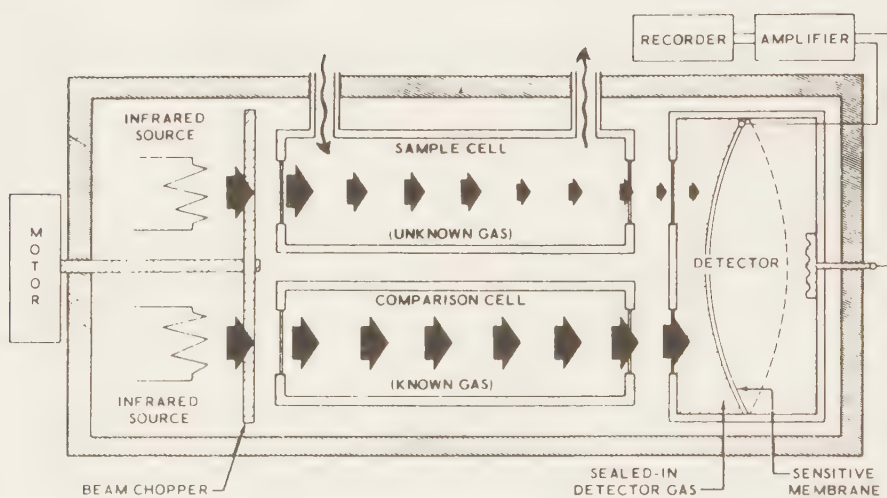


Figure 10. Infrared gas analyzer.

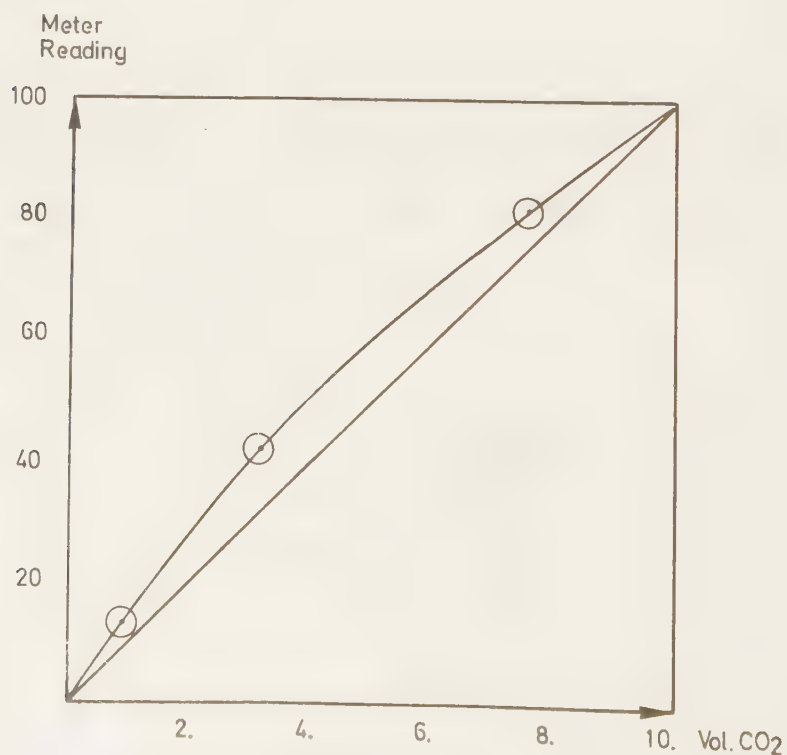


Figure 11. Gas calibration diagram.

6. METHOD OF INVESTIGATION

The three packings, plastic Intalox saddles, Tellerettes and Plasdek, were investigated so that both absorption and desorption data were achieved. Three different packing heights were also tested; 0.75 m, 1.5 m, and 3 m.

6.1 PACKING INSTALLATION

Three different methods of packing a tower randomly can be applied: dry shaken packing, dry unshaken packing, and wet packing. The efficiency of a packing is probably affected by the packing method, since there will be a different number of packing pieces per cubic meter in each case. In this investigation dry shaken packing of Intalox saddles and Tellerettes was used. The Plasdek, on the other hand, is manufactured in whole blocks of approximately 0.3 meters in height. The blocks are always placed upon each other with right angles between the flow channels (see fig. 1).

Table 2 indicates the number of packing pieces per cubic meter. Also shown are values supplied by manufacturers and various other investigators.

6.2 LIQUID AND GAS RATES

The mass transfer coefficient was determined for a wide range of irrigation rates; from 6 to 60 kg/m²·s. Most other investigators have used a range up to about 20 kg/m²·s. Cooper and Christl¹² have used approximately the same range. Their primary objective was to investigate the magnitude of the mass transfer in the loading-flooding region. The main objective of this investigation is to get the $k_L a$ -values at liquid rates encountered in environmental control, where for certain applications very high irrigation rates are used in order to get compact units. However, the range used here also covers most cases with chemical absorption where lower rates may be used.

In order to get good packing performance it is of great importance to have the packing well wetted. A measure of how well the packing is wetted is the wetting rate, which may be defined as²³

$$\eta = \frac{L}{\rho_L \cdot a} \quad (10)$$

If the packing is well wetted η should have a value greater than $3 \cdot 10^{-5} \text{ m}^3/\text{m} \cdot \text{s}$.

Calculations show that the irrigation rates used here all give good wetting. If η is too high ($> 11 \cdot 10^{-5}$) there may be unacceptably great liquid entrainment at high air velocities. This has not been observed although η had a value of $30 \cdot 10^{-5}$ at the highest liquid rate.

If the system $\text{CO}_2/\text{air-water}$ is entirely liquid-film controlled, the $k_L a$ -value is not affected by the gas rate. Any gas rate suitable for practical reasons can be chosen, provided it is high enough to prevent back-mixing of the gas. In this investigation three different superficial gas velocities have been used, 0.05 m/s, 0.1 m/s, and 0.2 m/s. The gas velocity of 0.05 m/s is probably somewhat low and back-mixing may occur. From a practical viewpoint it was difficult to have any velocity higher than 0.2 m/s because the carbon dioxide froze to ice in the gas cylinder valves. However flooding could occur at the highest irrigation rate.

6.3 EXPERIMENTAL PROCEDURE

Each test day, calibration diagrams for the p_{CO_2} -electrode and the gas analyzer were constructed. The calibration gases were measured at least three reproducible times. During the calibration the pumps were started and the system temperature raised to 298 K by the immersion heaters.

When the system temperature was adjusted and the calibration was accomplished, the gas and the liquid flows were adjusted to the desired values. Carbon dioxide was introduced into the air stream at the bottom of the absorber and the concentration was continuously registered by the gas analyzer. After a stable value of about 10 % carbon dioxide was achieved, the test was run for about 45 minutes, a time period proven to give equilibrium.

The absorber was then analysed; streams from the bottom being analysed before streams from the top. After about 10 minutes the desorber streams were analysed in the same manner. All analyses were then repeated in or-

der to give a second set of values. The total time for a run was about 90 minutes. Usually four runs were performed per test day.

Primarily a series of three gas rates and six liquid rates was tested. If any $k_L a$ -value obtained from this series differed in an anomalous way a new experiment was performed on the gas and liquid rate in question. The reproducibility was also checked by a series of new adjustments of gas and liquid flow rates. For each packing this was done with at least one packing height. The agreement was good among the series of runs performed.

Table 2

Number of packing pieces per cubic meter of packing

25 mm nominal size

	PP Intalox Saddles	PP Tellerettes
Manufacturer	58 000	36 000
Observed ($z = 0.75$ m)	72 000	41 000
Literature	53 500 ¹⁰	39 750 ²

7. EVOLUTION OF DATA

The calculations were performed on a digital computer. All mass flows and concentrations were converted into the desired units, the various constants were adjusted to the prevailing temperature, mass balances were established, and finally the $k_L a$ -value was calculated. The $k_L a$ -value was calculated by two different methods; one method which ignored the hydration and one which considered the hydration.

7.1 THE MASS BALANCE

The mass balance is established by comparing the amount of carbon dioxide transported to the liquid phase with the amount transported from the gas phase. (Reverse direction in desorption.)

$$\frac{\frac{L}{M_L} \cdot (x_1 - x_2)}{\frac{G_1}{M_{G_1}} \cdot y_1 - \frac{G_2}{M_{G_2}} \cdot y_2} = 1 \pm e \quad (11)$$

It is assumed that the liquid flow is constant throughout the tower. The entering and leaving gasflows are calculated as dry flows, since the gas samples were dried before they were analysed.

In experiments where the change in both liquid and gas composition was substantial the difference, e , in the mass balance was always below 10 % and was often as small as 1 %. However, when the change in composition of either the gas or the liquid stream was small the mass balance could show a significant difference. This occurred when a high gas flow rate was combined with a low liquid flow rate or when a low gas flow rate was combined with a high liquid flow rate.

From the mass balances it is concluded that each pair of analyses is very accurate, provided that there is a substantial change in carbon dioxide content between the top and the bottom of the tower.

7.2 CALCULATION OF THE MASS TRANSFER COEFFICIENT

To calculate the $k_L a$ -value the carbon dioxide concentration of three of the four streams from the tower must be known. When there is no difference in the mass balance the $k_L a$ -value can be calculated from either one gas analysis and two liquid analyses, or from two gas analyses and one liquid analysis. However, when the difference is significant the most reliable analyses should be used. The carbon dioxide content of the gas phase is known with highest accuracy at the bottom of the towers. This is firstly due to the fact that the gas composition here is homogeneous over the cross-sectional area. Secondly at this point in the desorption tower the concentration is the same as in normal atmospheric air and in the absorption tower the concentration is the highest in the system and therefore can be analysed with the best possible accuracy.

All liquid samples, irrespective of whether they were taken from the top or the bottom of the tower, have been analysed with the same thoroughness. The inhomogeneity of liquid phase carbon dioxide content, which may exist at the bottom of the tower, is diminished by the relatively large collecting area which is situated at two different points. All $k_L a$ -calculations therefore could be done, without losing any accuracy, by using the two bottom analyses and one top analysis. The top analysis used depended on which stream that had the greatest change in composition across the tower. This means that at high gas rates and low liquid rates the two liquid analyses are used, together with the bottom gas analysis, to calculate the $k_L a$ -value, while at low gas rates and high liquid rates the two gas analyses are used together with the bottom liquid analysis.

In order to simplify the computer program, used for the calculations only equations containing liquid concentrations were used. Therefore when the gas analyses were to be used the top liquid concentration was simply adjusted to comply with the mass balance.

7.2.1 Ignoring the Hydration

When a true physical mass transfer system with only liquid film resistance is used, the following relationship for calculating the $k_L a$ -value can be derived:

$$k_L a = \frac{L}{Z \cdot M_L \cdot \rho_m} \int_{x_2}^{x_1} \frac{dx}{(1-x) \cdot |(x^* - x)|} \quad (12)$$

Here subscript 1 denotes the maximum and subscript 2 the minimum concentration respectively prevailing at either end section of the tower, i.e. the same formula is applicable to both absorption and desorption.

The equation is solved numerically by Simpson's formula, using appropriate analyses to obtain the most representative $k_L a$ -value.

7.2.2 Considering the Hydration

When carbon dioxide is absorbed in water a hydration reaction takes place. This reaction may be taken into account by using a stepwise procedure when calculating the $k_L a$ -value. For this purpose the tower was divided into a number of incremental sections as illustrated in figure 12. In each of these sections the following balance is applicable

$$\frac{G_{n-1}}{M_{G_{n-1}}} \cdot y_{n-1} - \frac{G_n}{M_{G_n}} \cdot y_n = k_L a \cdot dz \cdot \rho_m \cdot (\bar{x}_n^* - \bar{x}_n^0) \quad (13)$$

where

$$\bar{x}_n^* = \frac{(y_n + y_{n-1})}{2} \cdot \frac{P \cdot He}{\rho_m} \quad (14)$$

$$\bar{x}_n^0 = \frac{x_n + x_{n+1}}{2} \quad (15)$$

Here x and y are the mole fractions of molecular carbon dioxide in liquid phase and in gas phase respectively.

When CO_2 reacts with water some of the dissolved carbon dioxide will, at equilibrium, be in the form of bicarbonate and carbonate ions. From the expression of the first stoichiometric ionization constant K_1 it follows:

$$|\text{HCO}_3| = K_1 \cdot |\text{CO}_2| / |\text{H}^+| \quad (16)$$

The total carbon dioxide content in the liquid, assuming that the carbonate content is negligible, is then:

$$|\text{CO}_2|_{\text{Tot}} = |\text{CO}_2| + |\text{HCO}_3| = |\text{CO}_2| \cdot \left(1 + \frac{K_1}{|\text{H}^+|}\right) \quad (17)$$

Consequently it follows that a mass balance over a section n will have the form:

$$\frac{G_{n-1}}{M_{G_{n-1}}} \cdot y_{n-1} - \frac{G_n}{M_{G_n}} \cdot y_n = \frac{L}{M_L} \cdot \left(x_n \cdot \left(1 + \frac{K_1}{|\text{H}^+|}\right)_n - x_{n+1} \cdot \left(1 + \frac{K_1}{|\text{H}^+|}\right)\right) \quad (18)$$

The proton concentration may be calculated from the electro-neutrality condition. However, measurements of pH and pCO_2 of water samples indicate that an excess of metal ions is present. The electro-neutrality condition therefore takes the following form:

$$|\text{H}^+| + |\text{excess Me-ions}| = |\text{HCO}_3| + |\text{OH}^-| \quad (19)$$

Consequently the proton concentration is calculated from:

$$|\text{H}^+| = - \frac{|\text{ex. Me-ions}|}{2} \pm \sqrt{\left(\frac{|\text{ex. Me-ions}|}{2}\right)^2 + K_w + K_1 \cdot |\text{CO}_2|} \quad (20)$$

Here K_w is the water dissociation constant.

The concentration of excess metal ions will not vary through the tower. It may be calculated from equation (19) using the analysis of water taken either from the top or the bottom of the tower.

From equations 13-15, 18 and 20 it is obvious that a stepwise calculation of the concentration profile over the tower may be performed. In this procedure the measured bottom concentrations are taken as starting values. A trial-and-error selected $k_L a$ -value is then used to calculate the appropriate top concentration. The $k_L a$ -value is adjusted until the calculated top concentration matches the experimental concentration.

7.3 ACCURACY IN DETERMINED MASS TRANSFER COEFFICIENTS

The maximum error in the determination of the $k_L a$ -value may be estimated very easily by using standard methods. Some illustrative calculations show that the maximum error in the $k_L a$ -values obtained by physical absorption is about $\pm 20 \%$, and that obtained by physical desorption is about $\pm 10 \%$ (see table 3). In this investigation values obtained by absorption generally have a maximum predicted error which is approximately two times that of the predicted error in values obtained by desorption.

One reason for this is apparent from figure 13. Here the operating lines and the equilibrium line are plotted for some runs, performed with Intalox saddles. It can be seen that the smallest driving force, calculated as the difference between an equilibrium value (given by the equilibrium line and hence considered correct) and a measured liquid concentration, is generally in the bottom of the towers. The bottom condition therefore influences the calculation of the $k_L a$ -value more than does the top condition. Unfortunately the bottom condition of the absorber is not as accurately established as the condition of the desorber. There are two reasons for this. Firstly, all liquid analyses have an estimated maximum error of about 1 percent of the measured value. This means that in cases, where the driving force is small compared to the liquid concentration the maximum error in the driving force is much greater than 1 percent. This is often the situation at the bottom of the absorber. The desorber, on the other hand, has a low concentration at the bottom and a driving force which is approximately equal to this liquid concentration. Thus the maximum error in the driving force will still be about 1 percent. The second reason is that the bottom gas composition of the desorber is established very exactly as that of the surrounding atmospheric air. ($y = 0.00030$).

An estimation of a statistic mean error is arrived at by calculating the standard deviation of each experimental series. Table 4 shows values so obtained. There is still a clear indication that the desorption measurements gave more reliable $k_L a$ -values than did the absorption measurements.

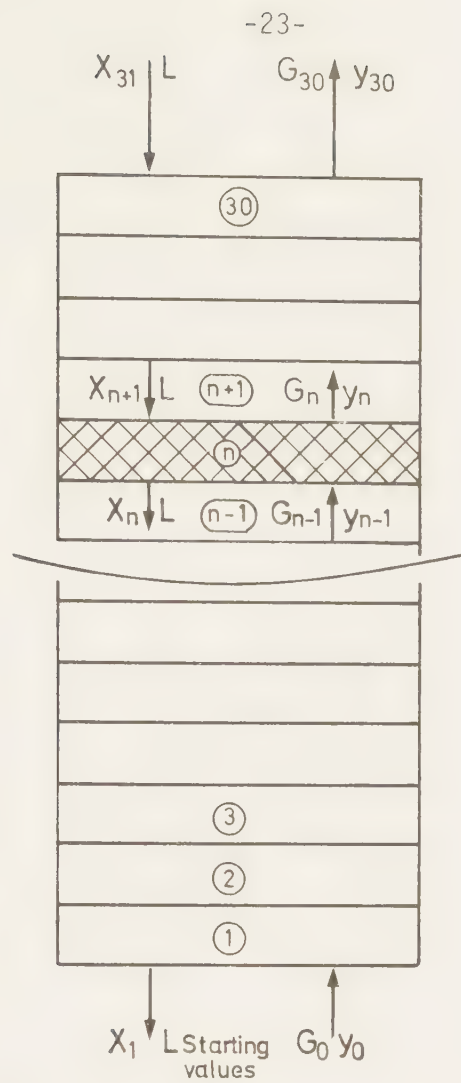


Figure 12. The tower divided into incremental sections.

Packing	Z m	Exp No	Absorption		Desorption	
			$k_L a$	Devi- ation	$k_L a$	Devi- ation
Plastic Intalox saddles	0.75	622	0.0124 ± 0.0013	15	0.0136 ± 0.0016	9
	1.5	152	0.0256 ± 0.0043	17	0.0412 ± 0.0042	10
Tellerettes	0.75	131	0.0068 ± 0.0011	15	0.0210 ± 0.0016	8
	1.5	351	0.0345 ± 0.0087	25	0.0365 ± 0.0062	17
		232	0.0345 ± 0.0055	16	0.0247 ± 0.0035	10
	3	61	0.0360 ± 0.0074	21	0.0174 ± 0.0023	15
Plasdek	0.75	151	0.0226 ± 0.0057	20	0.0566 ± 0.0062	11
	1.5	151	0.0435 ± 0.0061	14	0.0375 ± 0.0029	9
		261	0.0372 ± 0.0071	19	0.0353 ± 0.0046	13
	3	372	0.0129 ± 0.0022	17	0.0169 ± 0.0014	8
		381	0.0155 ± 0.0024	16	0.0152 ± 0.0011	8

Table 3. Maximum error in $k_L a$.

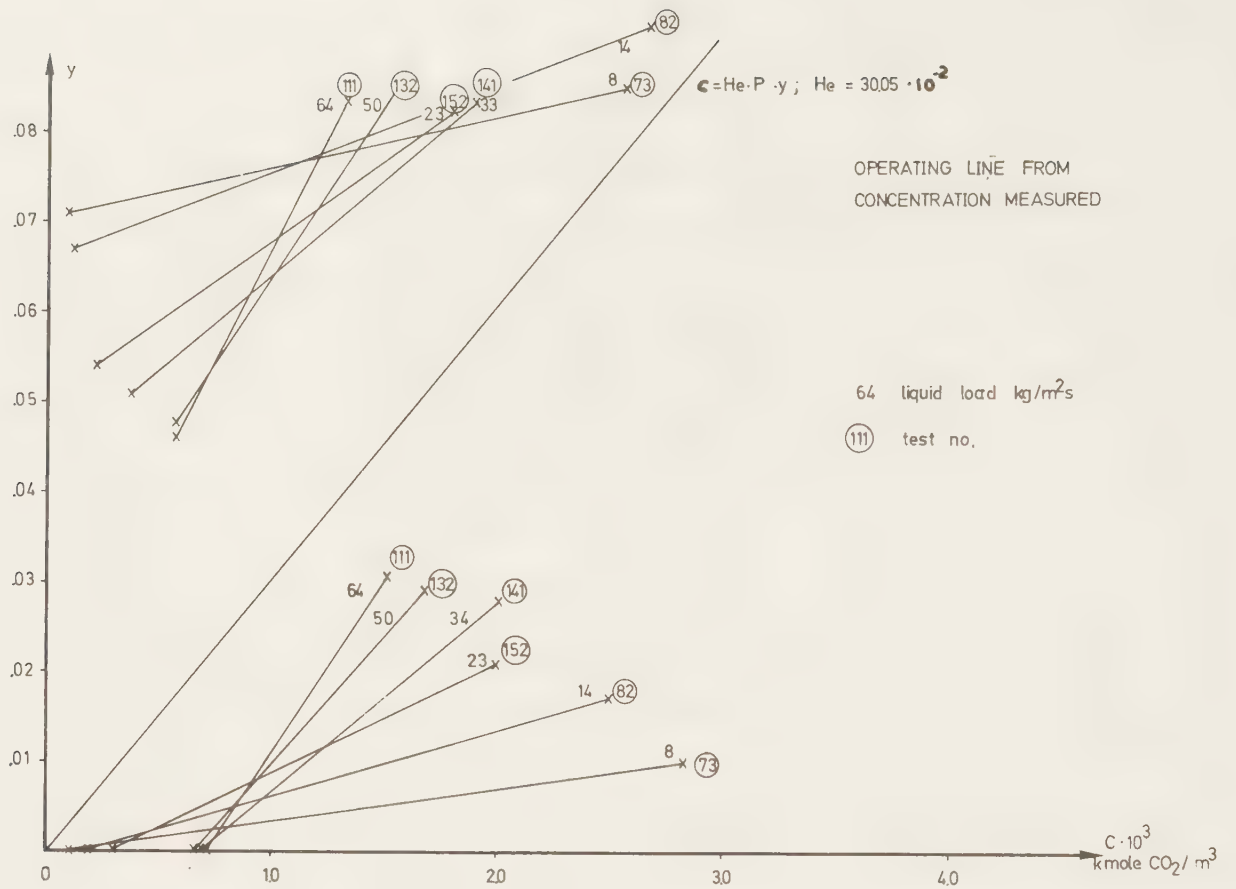


Figure 13. Operating and equilibrium lines.

Packing	Height	DESORPTION						ABSORPTION					
		G	L	$k_L a$	s	σ	n	G	L	$k_L a$	s	σ	n
Plastic Intalox saddles	0.75	0.054	13.77	0.0335	0.0011	3	4	0.048	13.76	0.0259	0.0025	10	4
		0.108	63.46	0.0830	0.0053	6	5	0.098	63.67	0.0623	0.0062	10	5
		0.204	22.47	0.0487	0.00024	1	4	0.185	22.51	0.0363	0.0043	12	4
	1.5	0.050	33.82	0.0438	0.0044	10	4	0.044	33.25	0.0314	0.00074	2	4
		0.100	33.82	0.0474	0.0013	3	4						
Tellerettes	0.75	0.054	63.70	0.0693	0.0051	7	4	0.048	65.45	0.0313	0.0034	11	4
		0.110	22.54	0.0446	0.00031	1	4	0.102	21.83	0.0218	0.0042	20	4
		0.206	22.55	0.0518	0.0038	7	4	0.186	34.52	0.0249	0.0024	10	4
	1.5	0.050	13.60	0.0172	0.00084	5	4	0.054	13.99	0.0195	0.0011	6	4
		0.099	7.53	0.0138	0.00054	4	5	0.097	7.14	0.0120	0.0017	14	5
		0.197	49.74	0.0355	0.0011	3	6	0.212	50.03	0.0336	0.0018	5	6
Plasdek	0.75	0.053	52.77	0.0679	0.0060	9	4	0.048	52.42	0.0382	0.0059	15	4
		0.111	22.26	0.0436	0.0030	7	4	0.100	21.93	0.0250	0.00058	2	3
		0.205	52.74	0.0820	0.0036	4	4	0.184	52.52	0.0376	0.0025	7	4
	1.5	0.050	31.83	0.0257	0.00083	3	4	0.055	33.54	0.0245	0.0015	6	4
		0.106	63.78	0.0461	0.0017	4	4	0.114	65.75	0.0473	0.0022	5	4
		0.194	49.26	0.0430	0.0028	6	4	0.216	50.11	0.0501	0.00067	1	4
	3	0.052	48.93	0.0166	0.0014	9	9	0.045	50.41	0.0189	0.00097	5	3
		Mean				5		Mean				8	

Table 4. Standard deviation of $k_L a$.

8. RESULTS

Liquid-side mass transfer coefficients from the experiments are gathered in figure 14-21. Here the $k_L a$ -values are plotted as a function of the liquid rate with the gas rate as a parameter. Each diagram presents average $k_L a$ -values for one packing at one packing height.

8.1 EFFECTS OF HYDRATION

A comparison between $k_L a$ -values calculated by the two different methods outlined in sections 7.2.1 and 7.2.2 shows no significant effect of hydration. As shown in figure 2 deionized water as used in this investigation in equilibrium with the surrounding atmosphere contains about half as much HCO_3^- as CO_2 . When absorbing CO_2 the fractional amount of HCO_3^- will decrease because of a decreasing pH (reaction (a) section 3). At pH = 5 (which was often the pH of the water at the highest carbon dioxide concentration encountered) the HCO_3^- -concentration at equilibrium is only 3 % of the carbon dioxide concentration. Likewise at the lowest carbon dioxide concentration encountered the pH was about 5.6 and the fractional HCO_3^- -concentration about 18 percent. Thus when carbon dioxide is absorbed in deionized water only 3-18 % takes part in the hydration reaction. It is therefore very likely that the two calculation methods would result in the same $k_L a$ -value.

However, it may also more stringently be shown that the hydration reaction has no influence on the absorption-desorption process; i.e. the reaction is a very slow reaction or a slow reaction. The conditions for these two types of reaction are given for instance by Danckwerts²⁴.

If the reaction is a very slow reaction only a negligible amount of the absorbed carbon dioxide reacts before leaving the absorber. The test condition is simply obtained by comparing the amount of carbon dioxide which has been absorbed to the amount which has reacted during the passage through the column. The amount of carbon dioxide absorbed is very easily calculated from a mass balance

$$\text{amount of } \text{CO}_2\text{-absorption} = \frac{L}{\rho_L \cdot z} (|\text{CO}_2|_1 - |\text{CO}_2|_2)$$

When carbon dioxide is absorbed by water a pseudo-first order equilibrium reaction takes place. A conservative test of whether the reaction has an influence on the process or not may be performed merely by taking the forward reaction into account. Furthermore, if it is assumed that the carbon dioxide content in the entire tower is constant and equal to the maximum concentration, the test is even more conservative. Thus the extent of reaction will be:

$$\text{extent of reaction} < \ell \cdot k_{\text{H}_2\text{O}} \cdot |\text{CO}_2|_{\text{max}}$$

The condition for the reaction to be classified as very slow then is

$$\frac{L}{\rho_L \cdot z} \cdot (|\text{CO}_2|_1 - |\text{CO}_2|_2) \gg \ell \cdot k_{\text{H}_2\text{O}} \cdot |\text{CO}_2|_{\text{max}} \quad (21)$$

In the case of desorption, carbon dioxide is produced by the reaction. It is now the reverse reaction which may influence the $k_L a$ -value. This is a second order reaction. A conservative test will be arrived at if the top concentrations are used and only the reverse reaction is considered. The analog of equation (21) for desorption thus will be

$$\frac{L}{\rho_L \cdot z} \cdot (|\text{CO}_2|_1 - |\text{CO}_2|_2) \gg \ell \cdot k_{\text{H}_2\text{O}}^{-1} \cdot |\text{H}^+| \cdot |\text{HCO}_3^-|_{\text{max}} \quad (22)$$

The CO_2 concentrations at the top and the bottom of the towers are known from the liquid analyses. Although these concentrations intrinsically also include the effect of reaction, they are believed to give an accurate measure of the amount of absorption and desorption respectively.

In table 5 the results of some tests on a very slow reaction are shown. Only the Tellerettes have been tested since this packing type seems to give the lowest mass transfer. Since no data on liquid hold-up have been reported in the literature, for the Tellerettes, a conservative value of 0.1 has been used. The tests indicate that the reaction may influence the process and hence the $k_L a$ -value. Clearly the extent of reaction is overestimated by the conservativity in the test, thus giving a measure of the maximum influence.

Measurements on the $\text{CO}_2\text{-H}_2\text{O}$ system may still give physical mass transfer data. The reaction may be classified as a slow reaction, which means that

there is no substantial reaction between the absorbed gas and a liquid component in the liquid film. A necessary condition for this is:

$$\frac{D}{k_L} \cdot r(A^*, B^0) \ll R \quad (23)$$

However, since the $k_L a$ -value is based on an interfacial liquid concentration in equilibrium with the gas phase concentration and a bulk equilibrium concentration, the reaction must be fast enough to keep the bulk at equilibrium, i.e. $A^* - A^0 \gg A^0 - A_e$. This is so when the following condition is fulfilled:

$$\lambda \cdot r(A^* - A_e, B^0) \gg k_L a \cdot (A^* - A^0) \quad (24)$$

If the bulk concentration deviates from the equilibrium value the transport coefficient will be underestimated.

In the case of a first order irreversible reaction the rate of reaction is expressed by:

$$r(\text{CO}_2^*, \text{H}_2\text{O}) = k_{\text{H}_2\text{O}} \cdot |\text{CO}_2^*| \quad (25)$$

The amount of absorption per unit film area may be expressed by the following relationship:

$$R = k_L \cdot (|\text{CO}_2^*| - |\text{CO}_2^0|) \quad (26)$$

Thus if there is to be no reaction in the liquid-film and a sufficiently fast reaction in the bulk-liquid to maintain the concentration at equilibrium, the following conditions must be fulfilled.

$$\frac{D_{\text{CO}_2}}{k_L} \cdot k_{\text{H}_2\text{O}} \cdot |\text{CO}_2^*| \ll k_L \cdot (|\text{CO}_2^*| - |\text{CO}_2^0|)$$

$$k_L a \cdot (|\text{CO}_2^*| - |\text{CO}_2^0|) \ll \lambda \cdot k_{\text{H}_2\text{O}} \cdot |\text{CO}_2^*|$$

or

$$\frac{D_{CO_2} \cdot k_{H_2O}}{k_L^2} \ll \frac{|CO_2^*| - |CO_2^0|}{|CO_2^*|} \ll \frac{\ell \cdot k_{H_2O}}{k_L a} \quad (27)$$

The desorption analog will be

$$\frac{D_{CO_2} \cdot k_{H_2O}^{-1} \cdot |H^+|}{k_L^2} \ll \frac{|CO_2^0| - |CO_2^*|}{|HCO_3^-|} \ll \frac{\ell \cdot k_{H_2O}^{-1} \cdot |H^+|}{k_L a} \quad (28)$$

This test should be performed on each part of the column. However, in the present study the absorption rate was generally smallest at the bottom of the column. Consequently if the conditions are fulfilled at the bottom they are also fulfilled in all other parts of the column.

To perform the test according to equations (27) and (28), the effective interfacial area and the liquid hold up of the packing must be known. Sahay and Sharma¹⁰ have reported values of interfacial area for 25 mm plastic Intalox saddles. Likewise values of liquid hold up may be found for instance in manuals from the manufacturer of the saddles¹. However, no determinations of these values for Tellerettes and Plasdek have yet been reported.

In table 6 some tests on plastic Intalox saddles are summarized. As seen in the table it is evident that the film-reaction may be disregarded. However the second test shows that the bulk concentration markedly deviates from equilibrium. Since all $k_L a$ -values obtained in this study are of the same order of magnitude, and since the effective interfacial area and liquid hold up do not vary to any great extent, it may be concluded that the hydration reaction tends to underestimate the $k_L a$ -value.

However since the tests show that there is no reaction in the film, while there is a rather large deviation from equilibrium in the bulk-liquid the reaction would more likely be classified as a very slow reaction. The test of a very slow reaction demonstrates that a maximum of 50 percent of the fraction of carbon dioxide, which is not at equilibrium,

reacts before leaving the column. As mentioned earlier the fraction of carbon dioxide which participates in the reaction varies between 3 and 18 percent through the tower. Thus as a mean value about 3 percent of the absorbed carbon dioxide reacts before leaving the tower. This will probably influence the $k_L a$ -value only to a minor degree, and will certainly not be detectable in values calculated by both of the methods. The carbon dioxide-water system therefore seems to be a physical mass transfer system and $k_L a$ -values can therefore be evaluated from equation (12).

8.2 EFFECTS OF LIQUID AND GAS RATES

When correlating liquid film data against liquid flow it is common to use an equation of the type

$$k_L a = \alpha(L)^\gamma \quad (29)$$

The exponent roughly has a value of 0.75-0.8 for most packings¹⁰. These γ -values have been reported by authors using liquid flow rates up to $20 \text{ kg/m}^2 \cdot \text{s}$. This investigation also shows that γ has a value of about 0.7-0.8 if L is smaller than $15 \text{ kg/m}^2 \cdot \text{s}$ (see table 7), with one exception, namely the Tellerettes. They seem to have a higher liquid rate dependency for absorption processes than other packings.

An increasing liquid flow rate results mainly in an increasing interfacial area and in a larger turbulence in the liquid film. Thus the mass transfer efficiency is increased. The increasing surface area, however, must also be accompanied by an increasing thickness of the liquid layer. The increase of the thickness becomes more pronounced as the packing becomes more completely wetted. Thus the γ -value of equation (29) should diminish as the liquid rate increases. Since in this investigation rather high liquid rates were used, and therefore the packings were very well wetted, the γ -value also was seen to decrease when L is greater than about $15 \text{ kg/m}^2 \cdot \text{s}$.

At the highest liquid rates used the $k_L a$ -values of the Tellerettes and the Plasdek tend to rise rapidly. As seen from pressure drop data (to be reported) the Tellerettes seem to work close to flooding condition when the highest flow rates are encountered. According to Cornell, Knapp and Fair²⁵ this will give a rapidly increasing mass transfer rate.

The Plasdek, on the other hand, shows no flooding, but only a small sudden increase in pressure drop at these rates. The liquid in this packing flows virtually as a film. When the downward velocity of this film is much higher than the upward velocity of the gas, the gas adjacent to the film will tend to move downwards, which gives a lower mass transfer rate. When the upward gas velocity is increased to a certain value all the gasfilm will move upwards and therefore the interfacial shear stresses and the pressure drop will increase. The mass transfer process will now change from being partially of a co-current nature to being fully counter-current which results in higher $k_L a$ -values. This explanation is also put forward by Cooper, Christl and Perry¹² in their investigation of Raschig rings at high flow rates.

From figures 14-21 a difference in $k_L a$ -values obtained by different gas velocities, especially at higher liquid rates, may also be noted. The main reason for this is probably back-mixing of the gas. A mixing of gas of different concentrations will diminish the driving force and thereby the mass transfer efficiency. Consequently gas velocities which are too low give lower $k_L a$ -values than gas velocities which are somewhat higher, especially at increasing liquid loads. The back-mixing effect will also increase with packing height, since the concentration differs more between the top and bottom of the tower. However, as indicated by the figures, it is believed that at a gas velocity of 0.2 m/s the back-mixing is of minor importance. Hence these $k_L a$ -values are representative of all gas velocities where no back-mixing and no flooding occur.

The $k_L a$ -values obtained with a packing depth of 3 meters do not show the same characteristics as the other $k_L a$ -values. This is probably due to the fact that the towers are operating more closely to equilibrium, i.e. these values contain more uncertainty. Consequently no data could be determined for the plastic Intalox saddles at this height. Likewise it was troublesome to get values at the gas velocity of 0.2 m/s for the others. However, a good many experiments were performed which yielded reproducible values. Thus the back-mixing effect may be confirmed. The data should only be used as a magnitude of order, and not directly for design purposes.

8.3 COMPARISON OF THE THREE INVESTIGATED PACKINGS

Liquid film mass transfer coefficients determined for the three investigated packings are compared in figures 22 and 23. It may be concluded that the mass transfer efficiency is relatively independent of packing type. If the packings are to be graded, the two figures show that the plastic Intalox saddles give the highest $k_L a$ -values, the Tellerettes the lowest, while $k_L a$ -values of the Plasdek lie between these. The deviation, and sometimes the order, among the packings may alter depending on process, height and liquid rate. For instance it is evident that the Intalox saddles have a superior mass transfer efficiency when used for absorption purposes. The efficiency of both the Tellerettes and the Plasdek is also more affected by different gas velocities; probably because in these packings the gas has a greater tendency to back-mix.

When comparing the packings it should be borne in mind that the Intalox saddles were more densely packed than the manufacturer recommends. The available transfer area was therefore greater, which presumably also enhanced the performance of the packing. However a denser packing also gives a greater pressure drop and a higher volumetric price than suggested by the manufacturer.

Any simple relationship valid for all parameters examined was not expected. As is readily observable, the three packings all operate differently. In a bed of Tellerettes liquid droplets are formed which splash from one packing piece to the next one and so on. The Plasdek consists of sheets arranged in a regular order. An annular type of flow exists with a liquid film on the sheets and the gas core between the sheets. The Intalox saddles are randomly packed and thus operate as Raschig rings, Pall rings, Berl saddles, and so forth.

Some liquid film data on 25 mm plastic Intalox saddles are reported in the literature by Sahay-Sharma¹⁰. Their values are plotted in figure 24 together with data from this investigation. The discrepancy may, as earlier mentioned, be due to the fact that in the present work a more densely packed column was used. From table 2 it may be seen that the number of packing pieces differed by about 20 %. The same difference

was also noted in the $k_L a$ -values. However, Sahay and Sharma used a column of only 0.2 m in diameter. Hence their values suffer more from maldistribution, which probably also contributes to the discrepancy.

8.4 COMPARISON OF OTHER PACKINGS

One of the main purposes of this investigation is to compare the investigated packings with other packings. Liquid film data of various commonly used packings have been gathered from the available literature. In table 8 some relevant references are listed. Data from these references are shown in figures 25-28, together with values obtained in the present study. In figure 25, $k_L a$ -values obtained with different 25 mm Raschig Rings are plotted. The agreement among different investigations is rather poor. A somewhat better agreement is achieved if all values are corrected to 25°C, as seen in figure 26. The desorption data in particular show improved agreement. The desorption data also point out that the $k_L a$ -value is relatively independent of whether carbon dioxide or oxygen is desorbed from water. This is to be expected, since the diffusion coefficients for the two gases in water are of the same order of magnitude.

In figure 27 $k_L a$ -values reported on Intalox saddles are compared. Due to different wetting properties the ceramic saddles should exhibit higher $k_L a$ -values than plastic ones do. No clear indication of this is evident from the diagram. However, two of the investigations on ceramic saddles yielded approximately the same $k_L a$ -values, but the third yielded somewhat lower $k_L a$ -values. Novella, Sancho, and Manford Doble used a column-to packing-diameter ratio substantially greater than 8, but Sahay and Sharma used a smaller column. Hence Sahay and Sharma's $k_L a$ -values may be lower because of greater maldistribution effects as was pointed out earlier. Furthermore, Sahay and Sharma also report a smaller number of packing pieces than is recommended by the manufacturer. A smaller number of packing pieces results in a smaller transfer area and thus in smaller $k_L a$ -values. Sahay and Sharma used about 12 % fewer packing pieces than for instance Manford Doble did.

Another variable which could explain some of the discrepancy is the absorption temperature. However not all investigators report what temperature they used. The $k_L a$ -values in the figure have therefore

not been corrected to the same temperature.

In figure 28 data for some miscellaneous packings are gathered. The data show that ceramic Berl saddles and stainless steel Pall rings have greater efficiency than plastic Intalox saddles and plastic Pall rings, possibly because of their different wetting properties. According to Sahay and Sharma¹⁰ all plastic packing should exhibit about the same efficiency. This was not confirmed by this study.

From figures 25 to 28 it may be concluded that packing efficiency is not influenced only by packing geometry. Other factors such as tower diameter, tower height, packing method, wetting properties, temperature and the physical properties of the test system also influence the efficiency to a comparable degree.

8.5 COMPARISON OF TRANSFER DATA OBTAINED BY ABSORPTION AND BY DESORPTION

In this investigation $k_L a$ -values were obtained both by absorption and by desorption of CO_2 in H_2O under equal experimental conditions. From figures 29, 30, and 31 it is evident that the direction of mass transfer, i.e. absorption and desorption respectively may influence the mass transfer efficiency. The influence seems to vary both with packing type and with packing height. At a packing height of 0.75 m the $k_L a$ -values of the Intalox saddles obtained by absorption is about 75 % of the $k_L a$ -values obtained by desorption. The Tellerettes and the Plasdek likewise have their efficiency diminished by about 50 %.

It is generally postulated in the literature that the same mass transfer efficiency is valid for both absorption and desorption. Sherwood and Holloway¹¹ for instance, by using Allen's data, showed that there was a close agreement between absorption and desorption data. More recent investigations, however, alter these findings. Thomas and Ray²⁶ made experiments on both absorption and desorption in static liquid pools. They concluded that the absorption process offers greater surface resistance to the mass transfer than does the desorption process. The same conclusion was arrived at by Gibbs and Himmelblau²⁷ from experiments performed under turbulent conditions. They also pointed out a dependency of the diffusion coefficient on concentration. As a result a larger $k_L a$ -value is obtained when desorbing CO_2 from water than when absorbing it. The difference should be at least five percent according to Gibbs and

Himmelblau²⁷. The difference obtained in the present investigation is considerably higher. This is probably due to an accumulation of impurities because the liquid was recirculated.

Figures 29, 30, and 31 show $k_L a$ -values obtained at a packing height of 0.75 m. When values from deeper packing sections are plotted the picture will be quite different. The Tellerettes and the Plasdek, as plotted in figures 32 and 33, now exhibit much less difference between data obtained by absorbing and desorbing the gas respectively. The Intalox saddles on the other hand still exhibit a difference, especially at higher liquid rates. The reason for this is not fully understood, although a possible explanation will be mentioned here.

As indicated before, the Tellerettes and the Plasdek seem to have a larger tendency to back-mix the gas than the Intalox saddles have. Superimposed on the back-mixing is a transport of CO_2 by eddy and molecular diffusion. This motion is directed towards regions with a lower carbon dioxide concentration, i.e. to the top of the absorption and to the bottom of the desorption tower. Since the driving force in most runs is smallest at the bottom, the $k_L a$ -values obtained by desorption will suffer more from deviation of the ideal plug flow condition. The effect of back-mixing will be diminished during absorption and enhanced during desorption due to the diffusion of CO_2 . Since concentration differences increase with increasing packing depths, back-mixing and diffusion also increase. As a result the difference between absorption and desorption will diminish with increasing packings depth.

A packed tower normally has a packing height larger than 1 meter. Also when a high packing section is needed redistributors are used at suitable distances. Therefore regardless of the foregoing discussion, it may be concluded (from diagram 32 and 33) that for most practical situations one need not be concerned whether the process is one of absorption or desorption.

8.6 EFFECTS OF PACKING HEIGHT ON THE $k_L a$ -VALUES. WALL FLOW

Many investigators^{9,10,11} have shown that the height of the packed section does not influence the $k_L a$ -value. However others, after careful examinations of reported data, have stated that there is such an influence.

For instance van Krevelen and Hoftijzer²⁸ reported a relationship of the following type:

$$k_L a \approx z^{-1/3} \quad (30)$$

Cornell et al²⁵ on the other hand recommended an exponent of -0.15.

In the present investigation three different packing heights were tested. As earlier mentioned there were great problems in obtaining values at the greatest packing height due to problems of saturation and back-mixing. However, reproducible results were achieved. In figures 34 to 36 $k_L a$ -values obtained at different heights are compared.

Two major effects of increasing packing depth can be assumed; channeling of water towards the tower wall, and back-mixing of gas elements with more differing concentration. From the figures it is apparent that the desorption mass transfer efficiency is influenced more by the packing depth than the absorption efficiency is. This is consistent with the larger back-mixing effect in desorption as discussed previously in paragraph 8.5. It may be concluded that when the Tellerettes and the Plasdek are used for absorption purposes there is no effect due to height provided redistributors are installed at suitable distances. The same conclusion probably applies also to the Intalox saddles. This, however, can not be demonstrated from present data, since values at the larger packing depth were only obtained at the lowest gas velocity.

On the other hand when the three packings are used for desorption purposes there is a significant effect from packing height. The height effect of the Intalox saddles may be correlated by using $z^{-0.2}$, with the exponent approximately constant for all liquid rates. The Tellerettes has a larger height dependency and the exponent here seems to vary from -0.4 to -1, depending on liquid rate. The height dependency of the Plasdek varies considerably with liquid rate; here the correlation method seems to be very inappropriate. However, an exponent of -0.5 to -1 may be attributed to the values of the Plasdek.

Since absorption and desorption exhibit quite different height dependencies, the main reason for the reduced efficiency is probably the back-

mixing. It is obvious that the channelling must be equal in the two towers. From previous investigations^{29,30} it is also concluded that the wall channelling has very little influence on the efficiency if the ratio of column diameter to packing diameter is larger than 8. This is because the maldistribution pattern is already established close to the top of the packing bed.

One method to cope with the height effect is to install redistributors at even intervals. Intervals of between 2 and 10 times the tower diameter are recommended¹⁹, depending on packing type. From the figures presented in this paragraph it follows that redistributors should be installed at an interval which is 2-3 times the column diameter, when the packings are used for desorption purposes. When they are used for absorption purposes the interval need only be from 5 to 7 times the column diameter. These intervals are based upon efficiency criteria only, and economic considerations may greatly alter them.

As was described earlier, a device to collect the wall flow was placed at the bottom of the column. A combined measure of self-distribution properties and wall channelling was thereby achieved. In table 9 relevant intervals of the wall flow are shown. The lower percentages of the interval were obtained at high liquid flow rates, while the higher percentages were obtained at low rates. Thus the fraction of liquid flowing on the walls diminishes at increasing liquid flow rates, a conclusion which agrees well with the results obtained by Porter and Tempelmann²⁹.

Since the "wall flow area" consists of 10 % of the total tower area, 10 % of the liquid should flow there if the distribution is good. If more is flowing down the wall the packing efficiency should be affected in a negative way. On the other hand, if less is flowing down the wall the self distribution properties of the packing are poor. As seen from the table the Intalox saddles and the Plasdek seem to distribute liquid towards the walls. This is true even at the lowest packing height. Though the wall flow increases somewhat with packing height, it may be concluded that the channeling immediately reduces the mass transfer efficiency. After this primary reduction the efficiency is relatively unaffected by the packing height. The Tellerettes on the other hand, seem to have poor self distribution properties. Thus the packing efficiency should increase with increasing packing depth because of improved wet-

ting of the packing. However, this effect is superceeded by an increasing back-mixing effect. When using the Tellerettes it is therefore essential to have a good liquid distributor.

Table 5

Tests of very slow reaction
Tellerettes

1. Absorption

Height m	Run No	L kg/s·m ²	$ CO_2 _1 = CO_2 _{\max}$ kmole/m ³	$ CO_2 _2$ kmole/m ³	amount of absorption	extent of reaction
0.75	11	7.14	$1.83 \cdot 10^{-3}$	$0.15 \cdot 10^{-3}$	$16 \cdot 10^{-6}$	$5 \cdot 10^{-6}$
	232	65.8	$1.41 \cdot 10^{-3}$	$0.85 \cdot 10^{-3}$	$49 \cdot 10^{-6}$	$4 \cdot 10^{-6}$
1.5	272	7.40	$2.70 \cdot 10^{-3}$	$0.19 \cdot 10^{-3}$	$12 \cdot 10^{-6}$	$7 \cdot 10^{-6}$
	221	64.82	$1.81 \cdot 10^{-3}$	$1.10 \cdot 10^{-3}$	$30 \cdot 10^{-6}$	$5 \cdot 10^{-6}$
3	41	13.79	$2.56 \cdot 10^{-3}$	$0.28 \cdot 10^{-3}$	$11 \cdot 10^{-6}$	$7 \cdot 10^{-6}$
	32	65.08	$2.32 \cdot 10^{-3}$	$1.37 \cdot 10^{-3}$	$20 \cdot 10^{-6}$	$6 \cdot 10^{-6}$

2. Desorption

Height m	Run No	L kg/m ² ·s	$ CO_2 _1$ kmole/m ³	$ CO_2 _2$ kmole/m ³	$ HCO_3^- _{\max}$ kmole/m ³	$ H^+ $ kmole/m ³	amount of absorption	extent of reaction
0.75	11	7.08	$2.58 \cdot 10^{-3}$	$0.35 \cdot 10^{-3}$	$0.55 \cdot 10^{-4}$	$2.08 \cdot 10^{-5}$	$21 \cdot 10^{-6}$	$6 \cdot 10^{-6}$
	232	64.23	$1.41 \cdot 10^{-3}$	$0.88 \cdot 10^{-3}$	$0.17 \cdot 10^{-3}$	$0.38 \cdot 10^{-5}$	$45 \cdot 10^{-6}$	$4 \cdot 10^{-6}$
1.5	272	7.46	$2.78 \cdot 10^{-3}$	$0.25 \cdot 10^{-3}$	$0.11 \cdot 10^{-3}$	$1.10 \cdot 10^{-5}$	$13 \cdot 10^{-6}$	$7 \cdot 10^{-6}$
	221	63.84	$1.80 \cdot 10^{-3}$	$1.13 \cdot 10^{-3}$	$0.65 \cdot 10^{-4}$	$1.23 \cdot 10^{-5}$	$28 \cdot 10^{-6}$	$5 \cdot 10^{-6}$
3	41	13.77	$2.68 \cdot 10^{-3}$	$0.26 \cdot 10^{-3}$	$0.69 \cdot 10^{-4}$	$1.74 \cdot 10^{-5}$	$11 \cdot 10^{-6}$	$7 \cdot 10^{-6}$
	32	63.84	$2.35 \cdot 10^{-3}$	$1.30 \cdot 10^{-3}$	$0.68 \cdot 10^{-4}$	$1.55 \cdot 10^{-5}$	$22 \cdot 10^{-6}$	$6 \cdot 10^{-6}$

Table 6

Tests of slow reaction
Plastic Intalox saddles (height: 0.75 m)

A. Absorption

Exp No	v m/s	L kg/m ² ·s	$k_L a$ 1/s	a m ² /m ³	ℓ	$ CO_2^* $ kmole/m ³	$ CO_2^0 $ kmole/m ³	$\frac{D_{CO_2} \cdot k_{H_2O}}{k_L^2}$	$\frac{ CO_2^* - CO_2^0 }{ CO_2^* }$	$\frac{k_{H_2O}}{k_L a}$
622	0.05	7.2	0.0102	125	0.045	$2.84 \cdot 10^{-3}$	$1.79 \cdot 10^{-3}$	$7.6 \cdot 10^{-3}$	0.37	0.11
242	0.05	13.6	0.0261	170	0.067	$2.91 \cdot 10^{-3}$	$2.20 \cdot 10^{-3}$	$2.2 \cdot 10^{-3}$	0.24	0.07
251	0.1	7.4	0.0158	125	0.045	$2.88 \cdot 10^{-3}$	$2.30 \cdot 10^{-3}$	$3.2 \cdot 10^{-3}$	0.20	0.07
262	0.1	13.8	0.0265	170	0.067	$2.91 \cdot 10^{-3}$	$2.15 \cdot 10^{-3}$	$2.1 \cdot 10^{-3}$	0.26	0.07
361	0.19	7.4	0.0187	125	0.045	$2.88 \cdot 10^{-3}$	$2.38 \cdot 10^{-3}$	$2.3 \cdot 10^{-3}$	0.18	0.06
352	0.19	13.8	0.0279	170	0.067	$2.82 \cdot 10^{-3}$	$2.20 \cdot 10^{-3}$	$1.9 \cdot 10^{-3}$	0.22	0.06

B. Desorption

Exp No	v m/s	L kg/m ² ·s	$k_L a$ 1/s	a m ² /m ³	ℓ	$ CO_2^0 $ kmole/m ³	$ HCO_3^- $ kmole/m ³	$ H^+ $ kmole/m ³	$\frac{D_{CO_2} \cdot k_{H_2O}^+ H^+ }{k_L^2}$	$\frac{ CO_2^0 - CO_2^* }{ CO_2^0 }$	$\frac{\ell \cdot k_{H_2O}^+ H^+ }{k_L a}$
471	0.05	7.14	0.0194	125	0.045	$0.434 \cdot 10^{-3}$	$0.173 \cdot 10^{-3}$	$1.12 \cdot 10^{-6}$	$5.3 \cdot 10^{-3}$	2.45	0.15
482	0.05	14.0	0.0322	170	0.067	$0.601 \cdot 10^{-3}$	$0.168 \cdot 10^{-3}$	$1.55 \cdot 10^{-6}$	$5.0 \cdot 10^{-3}$	3.51	0.18
371	0.1	7.4	0.0215	125	0.045	$0.338 \cdot 10^{-3}$	$0.396 \cdot 10^{-3}$	$3.80 \cdot 10^{-7}$	$1.5 \cdot 10^{-3}$	0.82	0.05
381	0.1	14.1	0.0354	170	0.067	$0.436 \cdot 10^{-3}$	$0.372 \cdot 10^{-3}$	$5.25 \cdot 10^{-7}$	$1.4 \cdot 10^{-3}$	1.14	0.06
361	0.2	7.4	0.0199	125	0.045	$0.337 \cdot 10^{-3}$	$0.498 \cdot 10^{-3}$	$3.02 \cdot 10^{-7}$	$1.4 \cdot 10^{-3}$	0.65	0.04
351	0.2	13.9	0.0337	170	0.067	$0.457 \cdot 10^{-3}$	$0.415 \cdot 10^{-3}$	$4.90 \cdot 10^{-7}$	$1.4 \cdot 10^{-3}$	1.07	0.08

$D_{CO_2} (0.1 \text{ MPa}, 25^\circ\text{C}) = 1.96 \cdot 10^{-9} \text{ m}^2/\text{s}^{18}$, $k_{H_2O} = 0.026 \text{ 1/s}$, $k_{H_2O}^{-1} = 58600 \frac{\text{m}^3}{\text{kmole} \cdot \text{s}}$

In diagram 14-21 the following notation is used:

- $v=0.05$ m/s
- ▽ $v=0.1$ m/s
- $v=0.2$ m/s

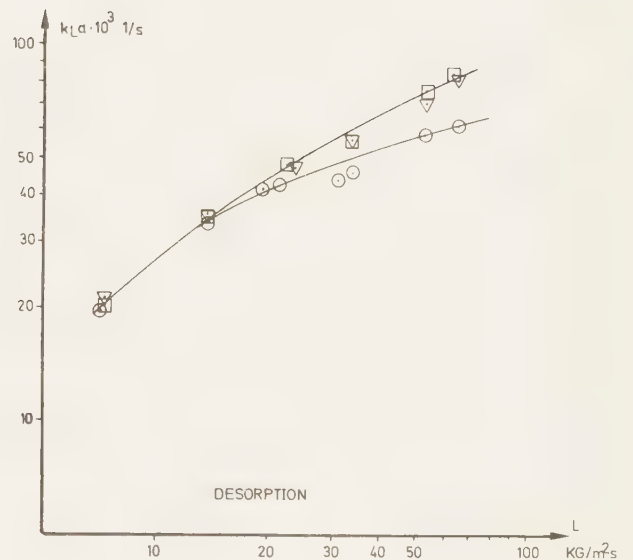
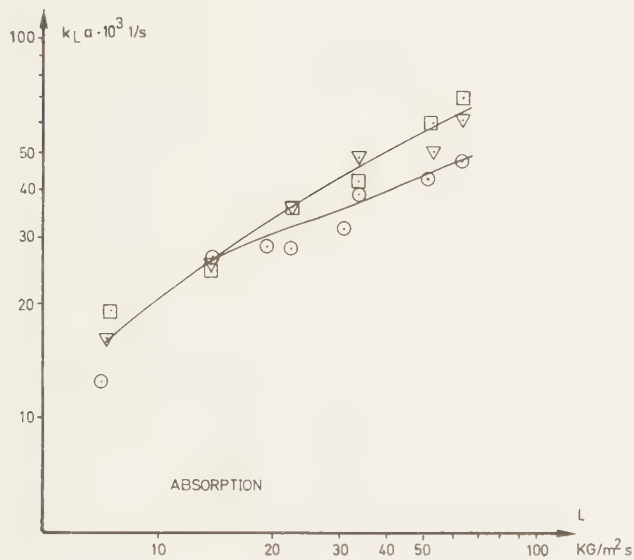


Figure 14. $k_L a$ -values obtained with Intalox saddles, $z=0.75$ m.

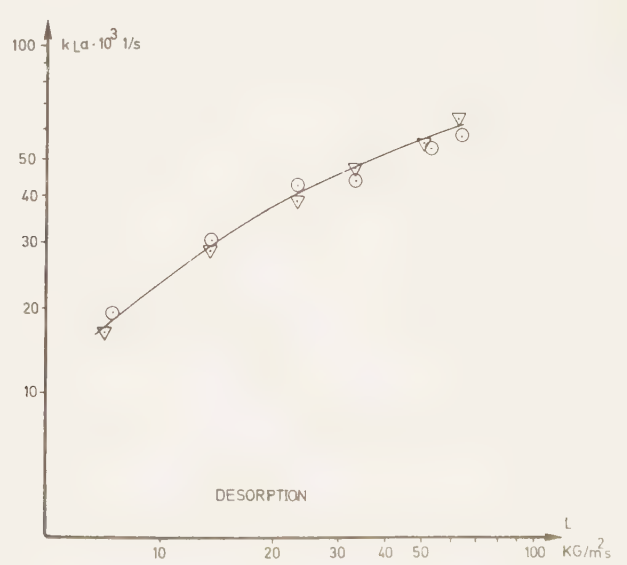
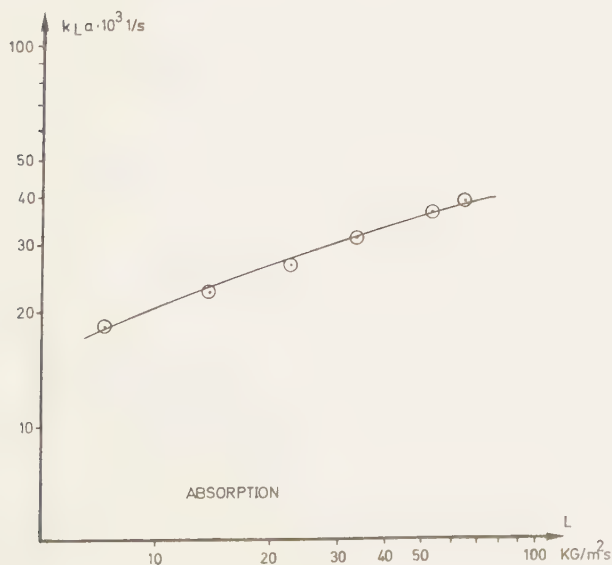


Figure 15. $k_L a$ -values obtained with Intalox saddles, $z=1.5$ m.

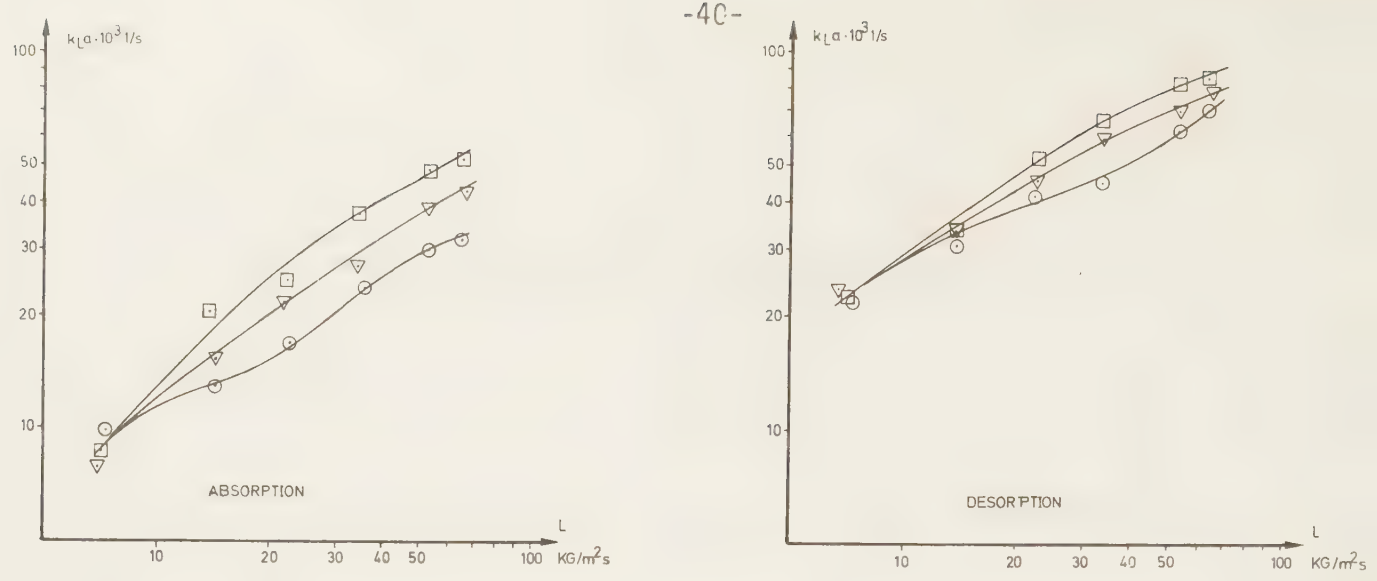


Figure 16. $k_L a$ -values obtained with Tellerettes, $z=0.75 \text{ m}$.

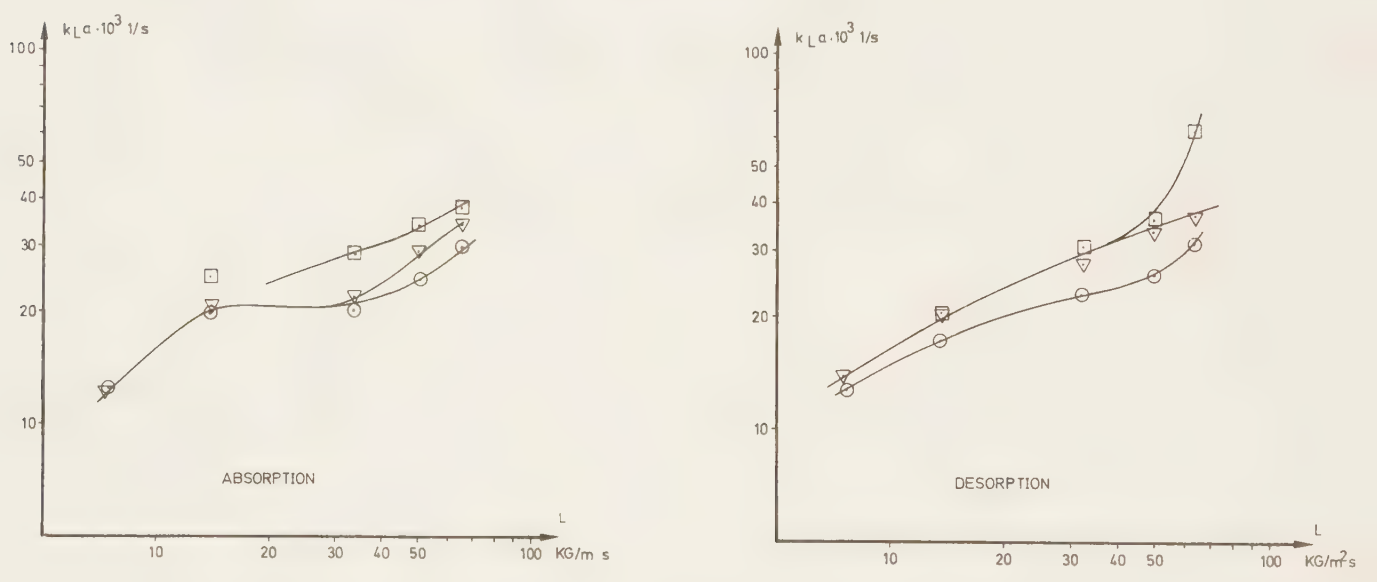


Figure 17. $k_L a$ -values obtained with Tellerettes, $z=1.5 \text{ m}$.

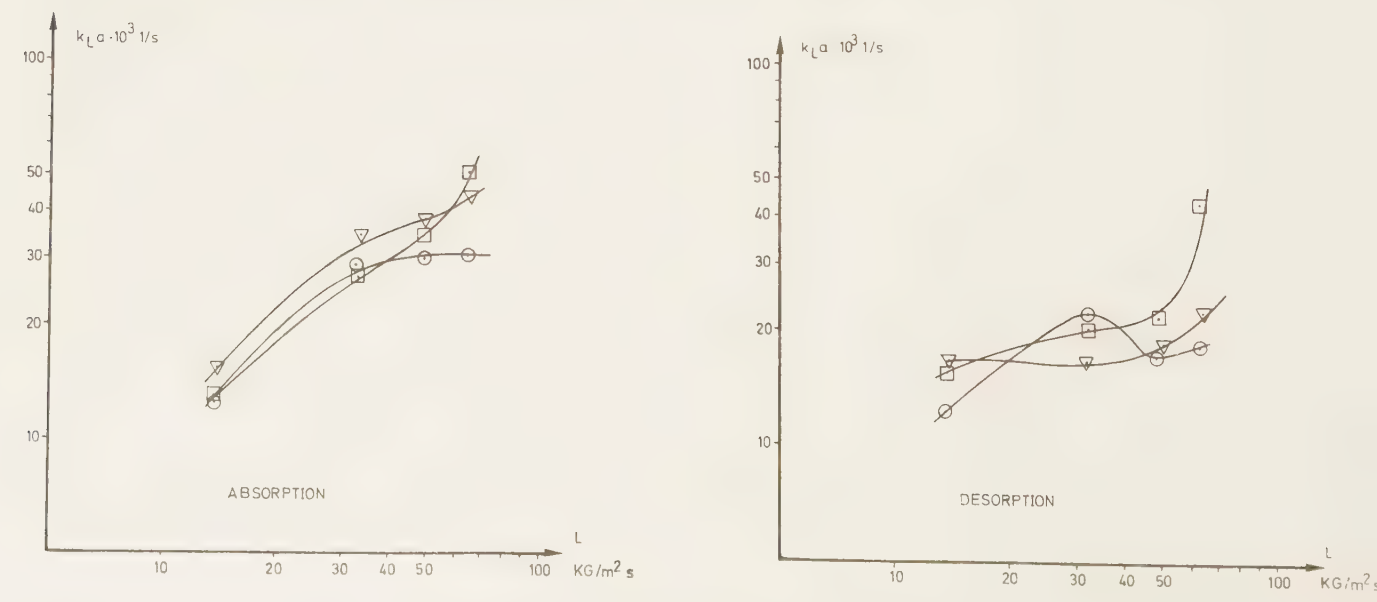


Figure 18. $k_L a$ -values obtained with Tellerettes, $z=3 \text{ m}$.

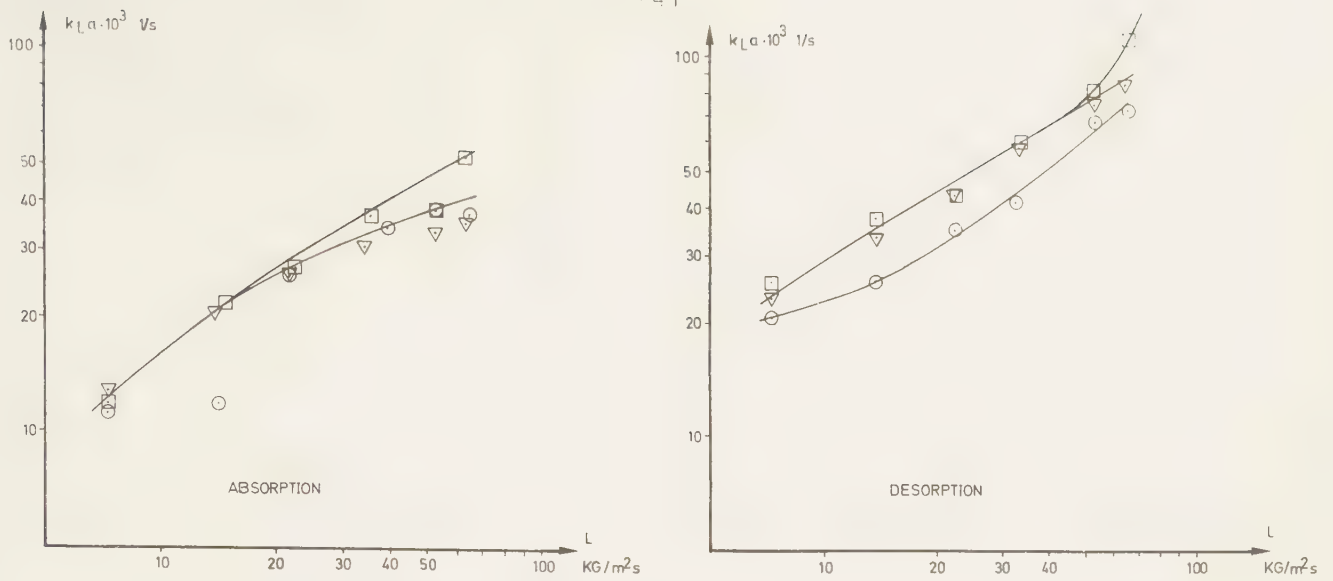


Figure 19. $k_L a$ -values obtained with Plasdek, $z=0.75 \text{ m}$.

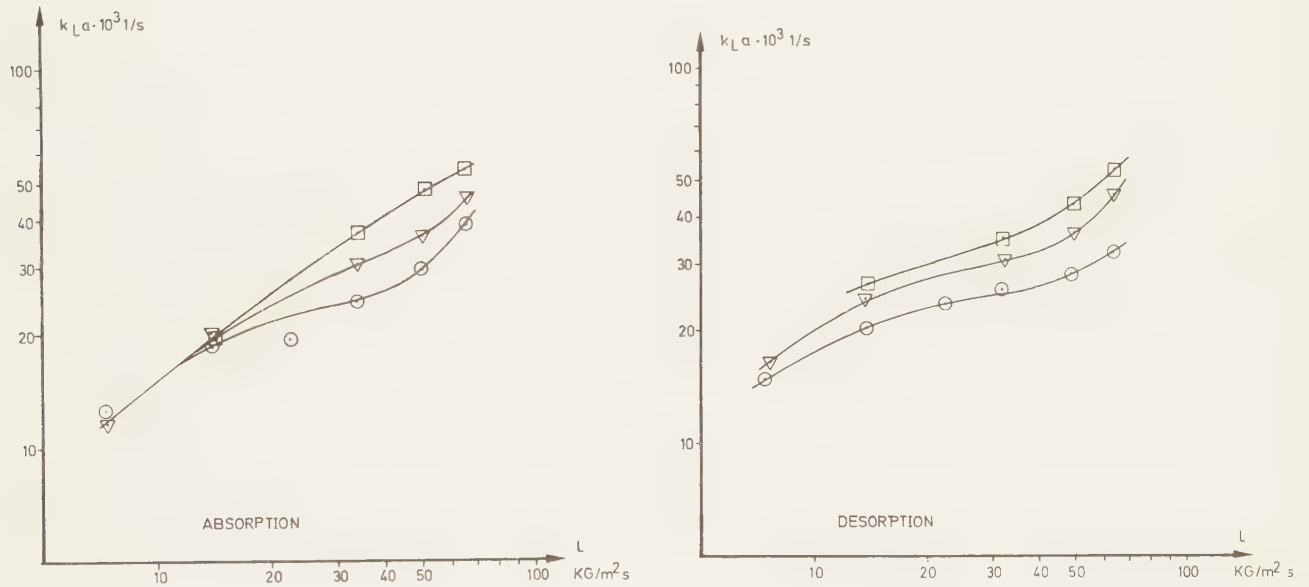


Figure 20. $k_L a$ -values obtained with Plasdek, $z=1.5 \text{ m}$.

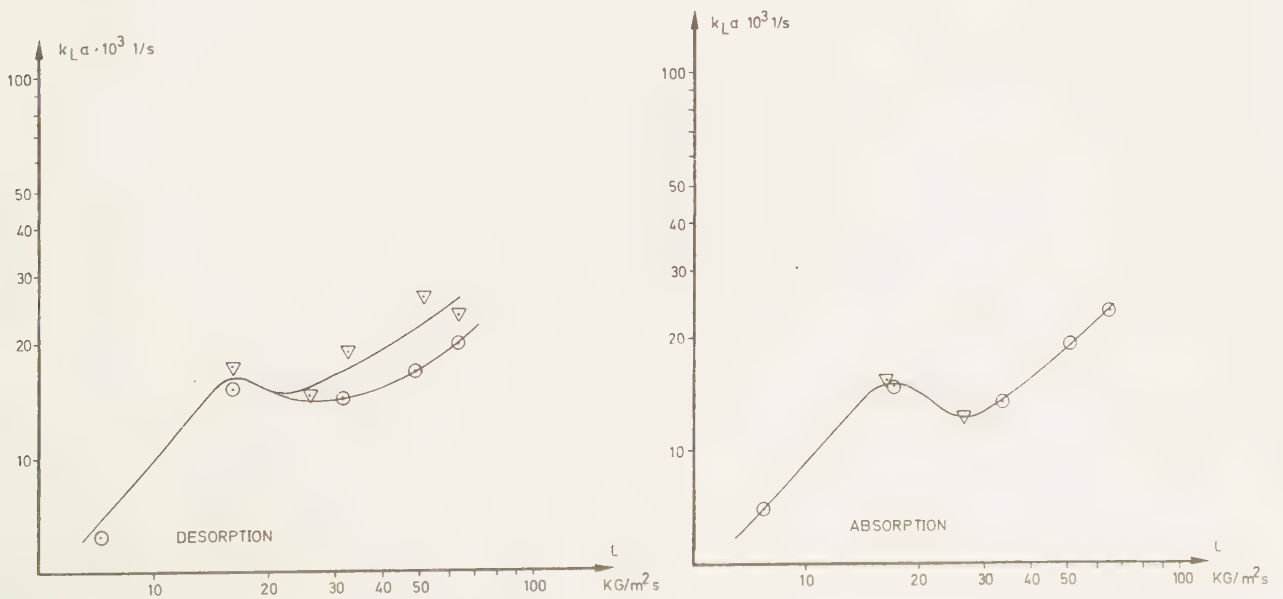


Figure 21. $k_L a$ -values obtained with Plasdek, $z=3 \text{ m}$.

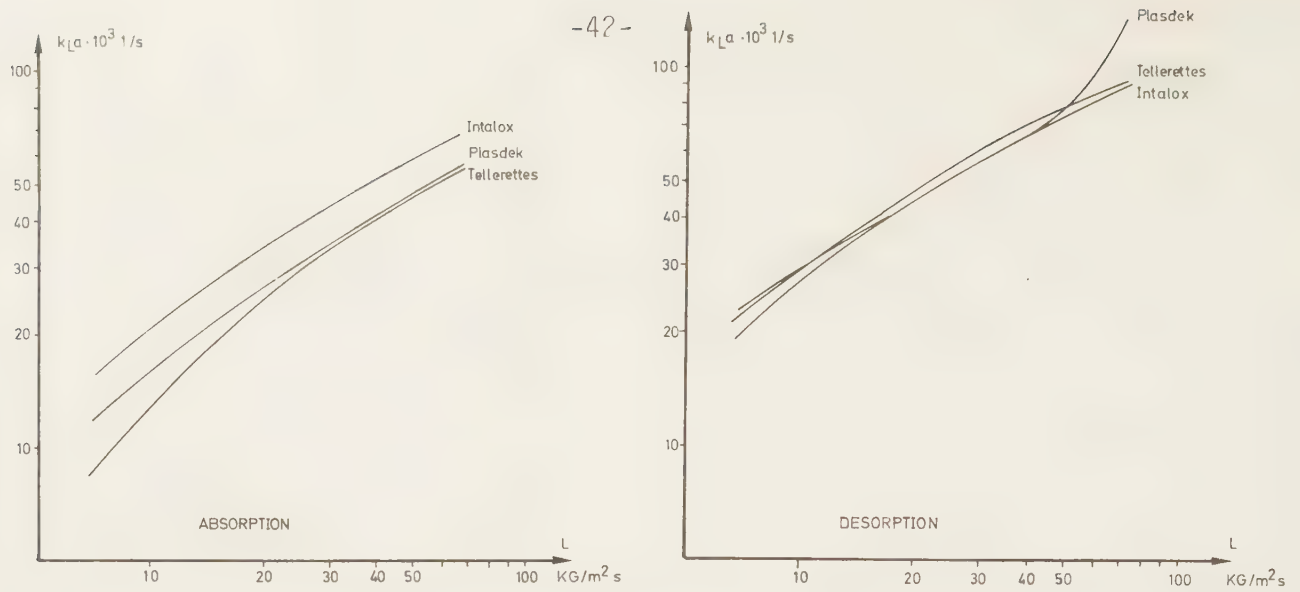


Figure 22. Comparison of $k_L a$ -values from the three packings, $z=0.75$ m.

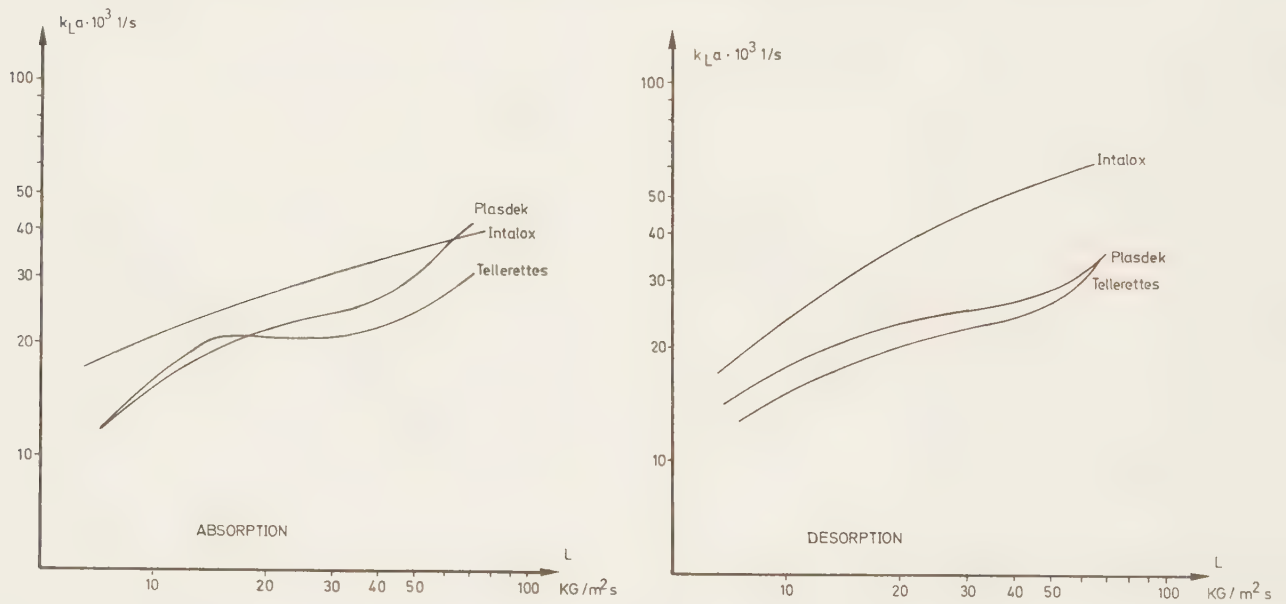


Figure 23. Comparison of $k_L a$ -values from the three packings, $z=1.5$ m.

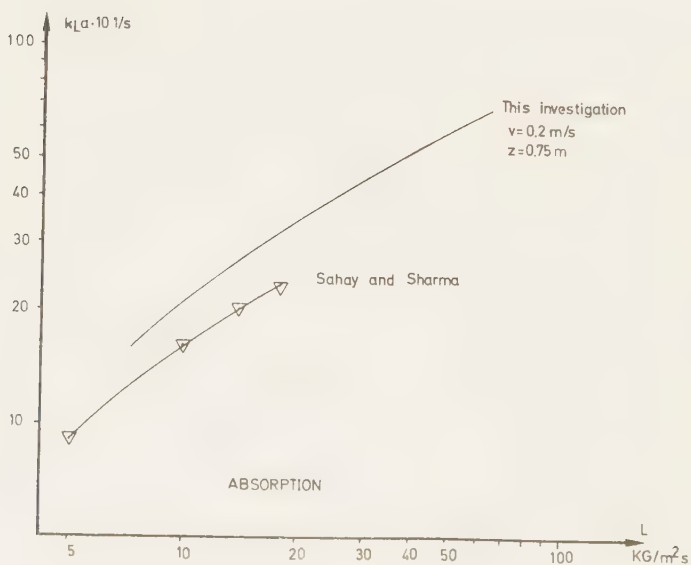


Figure 24. Comparison with results of Sahay and Sharma

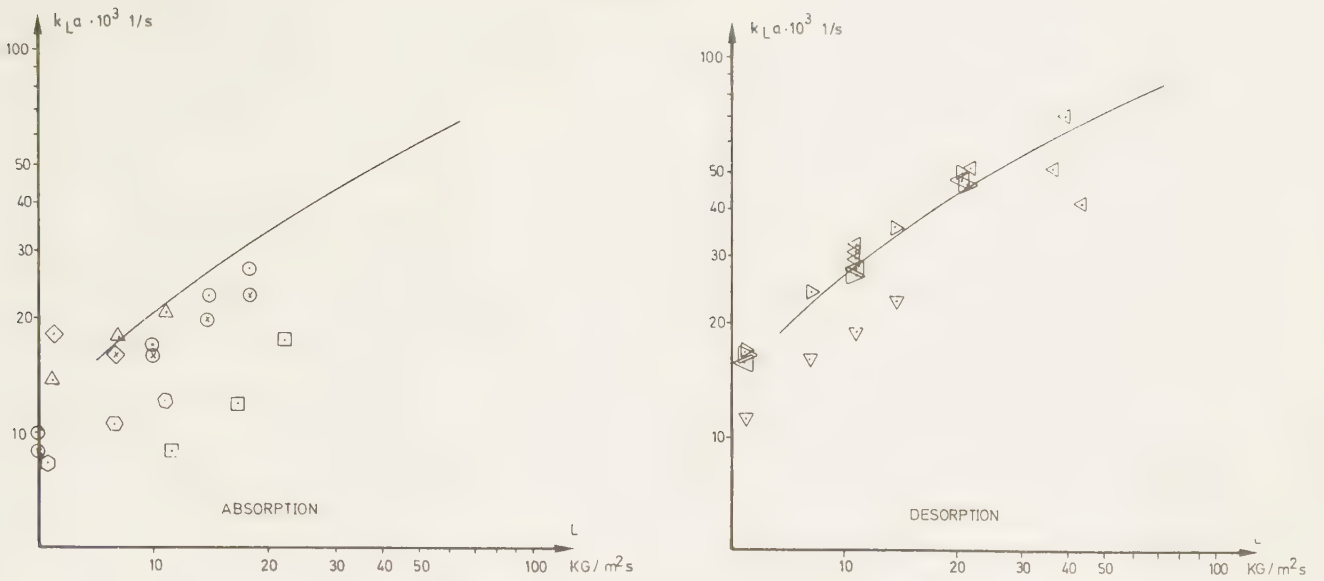


Figure 25. Literature data, Raschig rings.

Notations in figures 25 and 26

- △ Ceramic RR⁸
- Ceramic RR¹⁰
- ⊗ PVC RR¹⁰
- ⊠ Ceramic RR¹⁵
- ◇ Carbon RR¹⁶
- ⊙ Porcelain RR³
- Ceramic RR¹³

- ◁ Ceramic RR¹¹
 - ▷ Ceramic RR⁶
 - ◀ Tile RR⁶
 - ▶ Tile RR⁷
 - ▽ Carbon RR¹⁴
- } O₂ from H₂O
- } CO₂ from H₂O
- This investigation

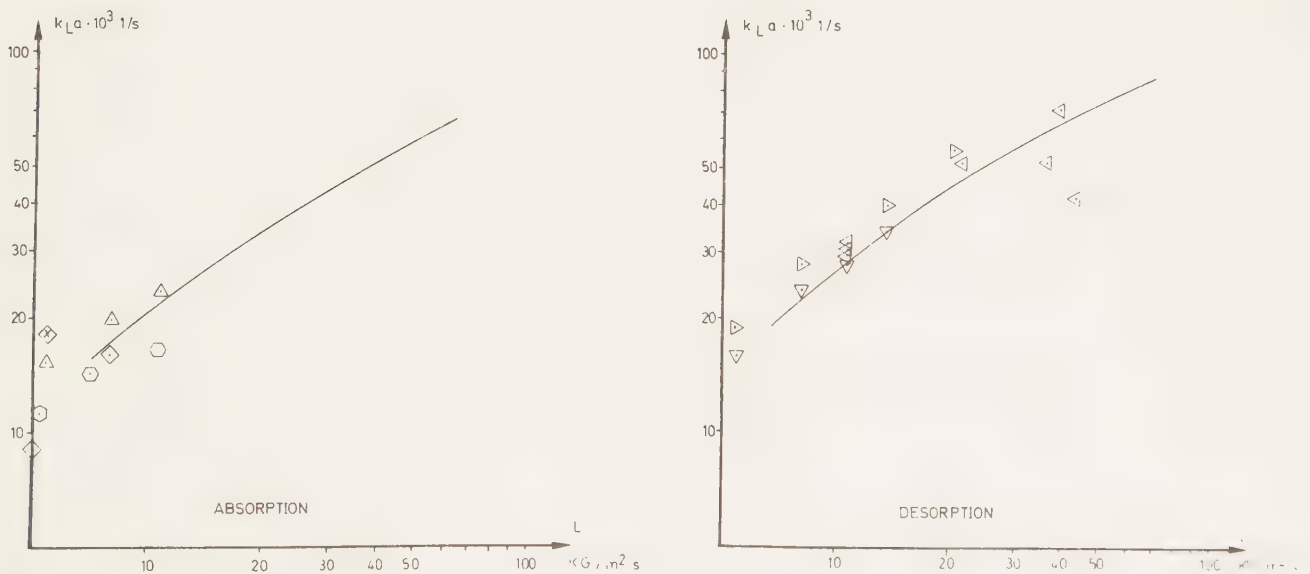


Figure 26. Literature data, Raschig rings corrected to 25 C

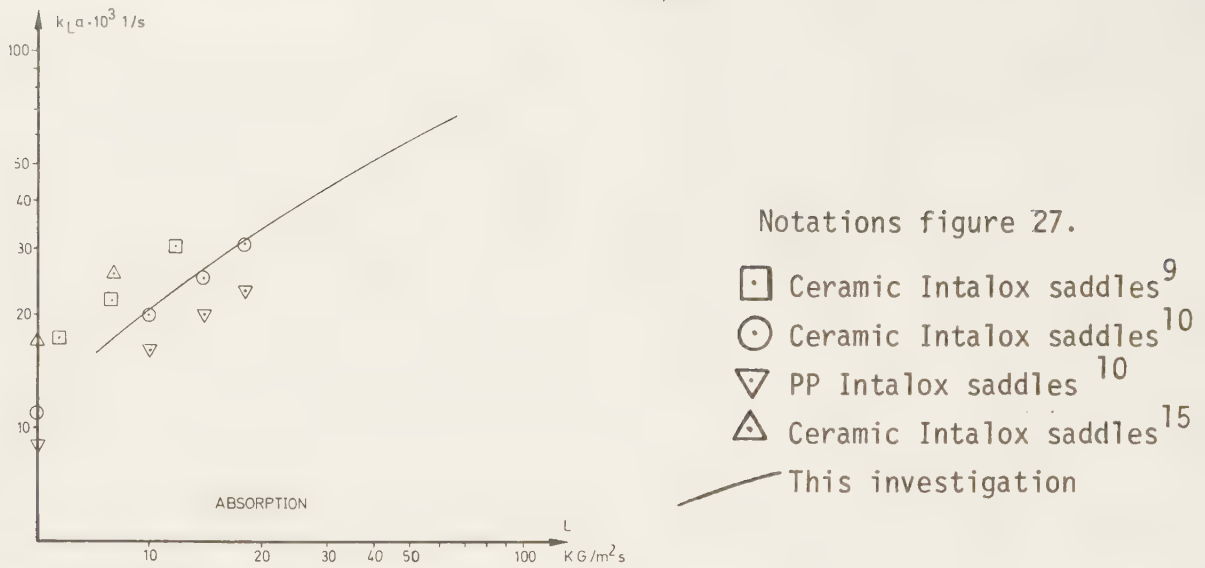


Figure 27. Literature data, Intalox saddles.

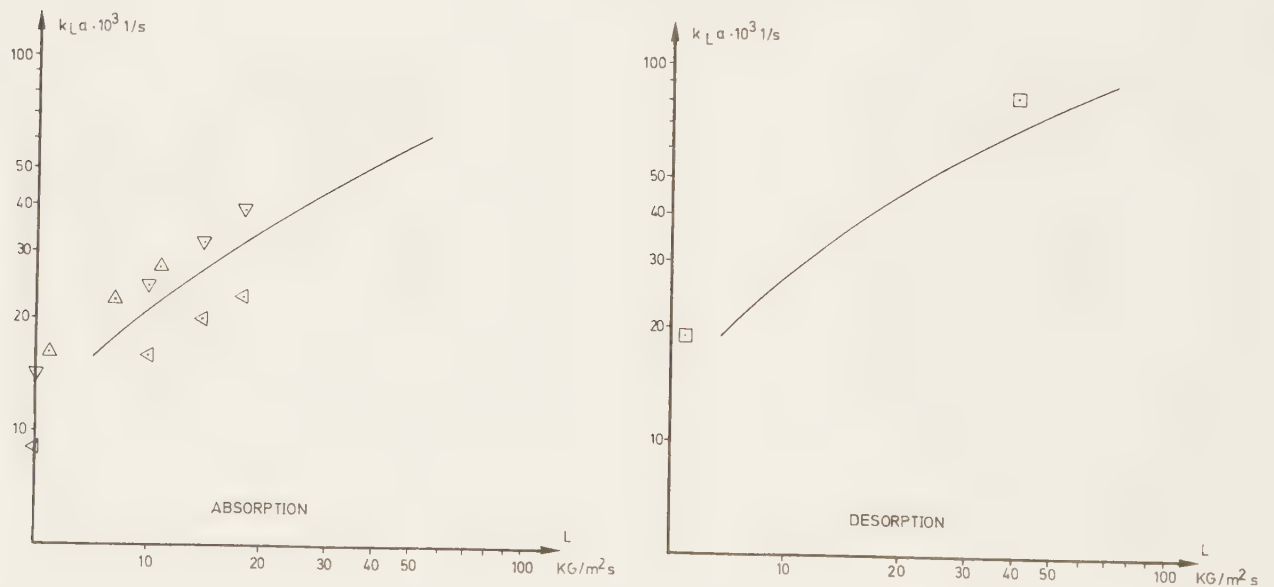


Figure 28. Literature data, miscellaneous packings.

Notations figure 28.

- Ceramic Berl saddles¹¹ O₂ from H₂O
- △ Ceramic Berl saddles⁸
- ▽ SS Pall rings¹⁰
- ◁ PP Pall rings¹⁰
- ◁ PP Intalox saddles¹⁰

— This investigation

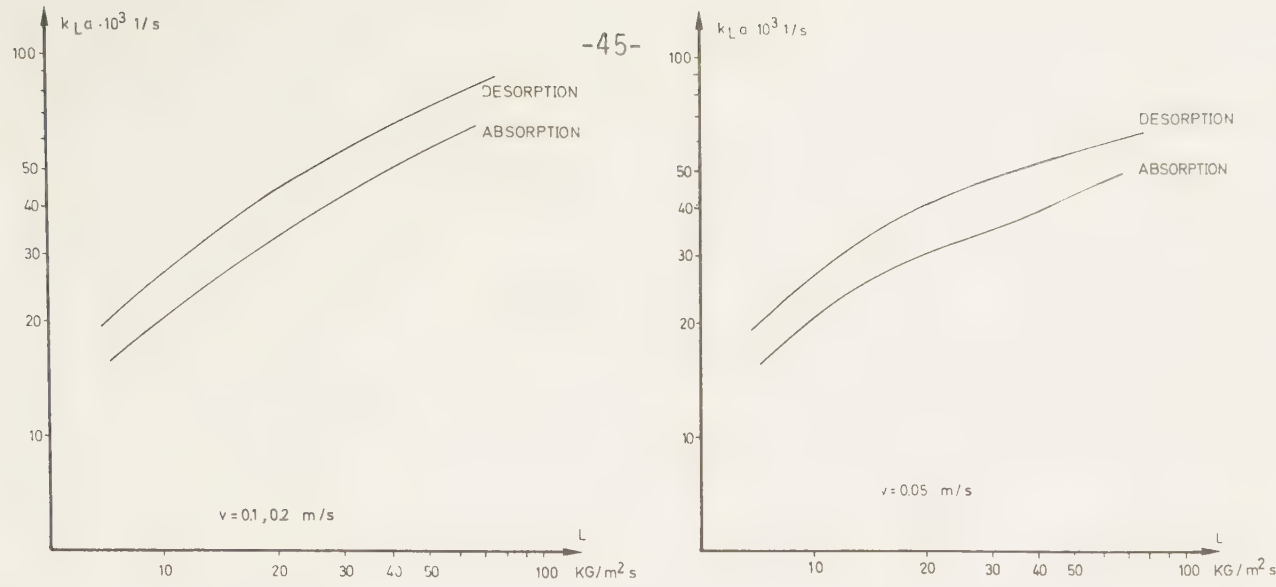


Figure 29. Effect of process, Intalox saddles at $z=0.75$ m.

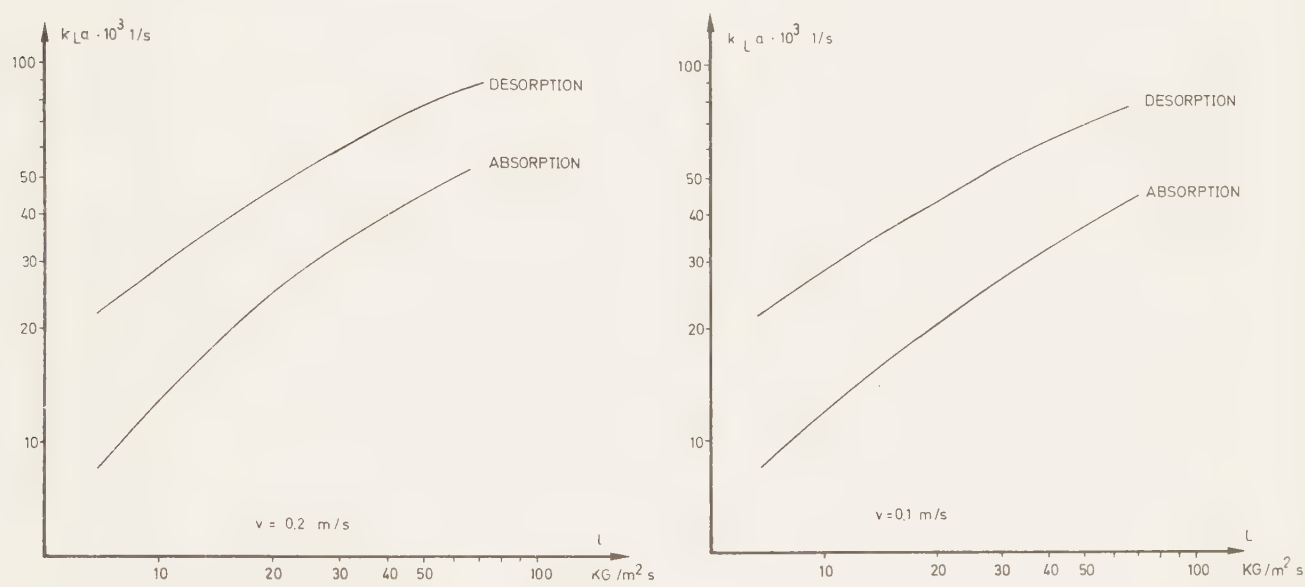


Figure 30. Effect of process, Tellerettes at $z=0.75$ m.

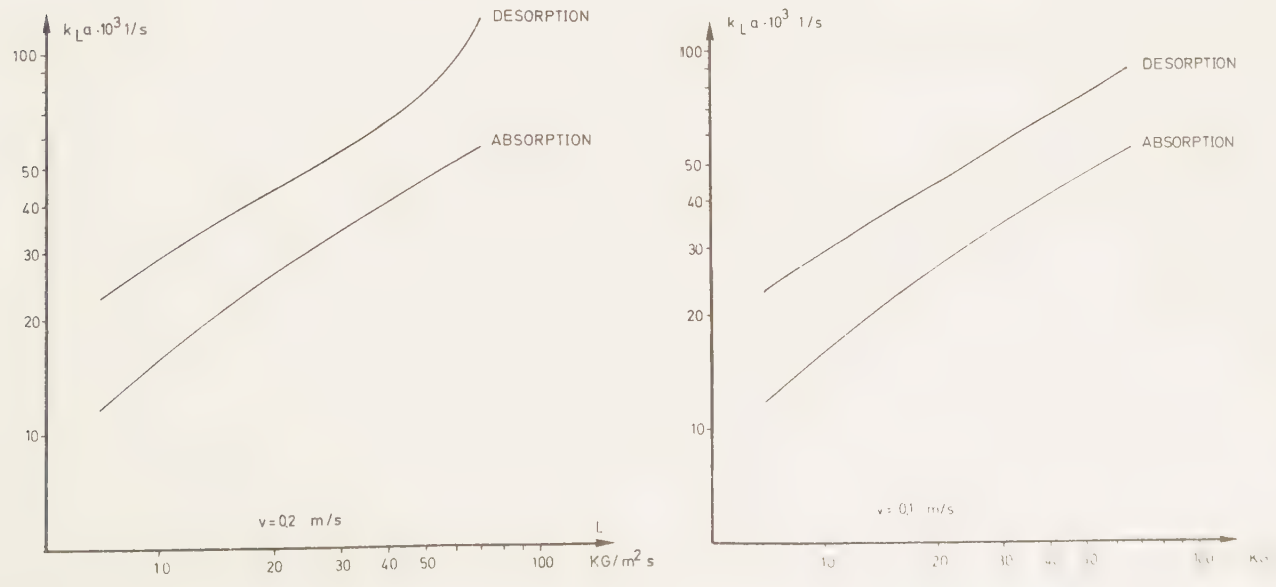


Figure 31. Effect of process, Plasdek at $z=0.75$ m.

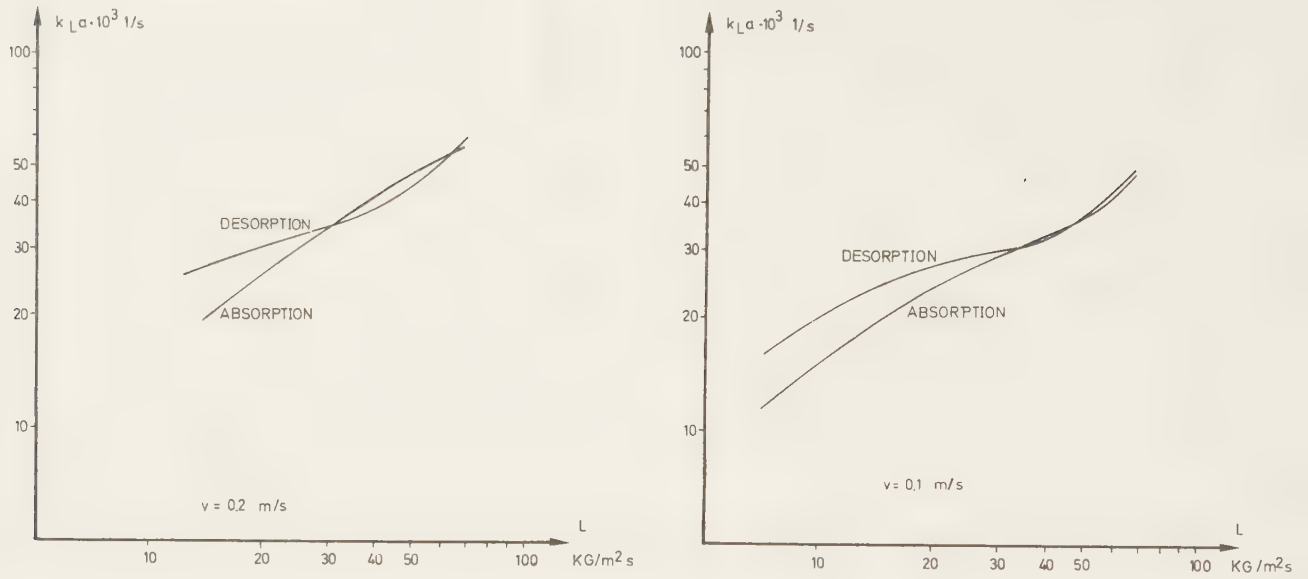


Figure 32. Effect of process, Plasdek at $z=1.5 \text{ m}$.

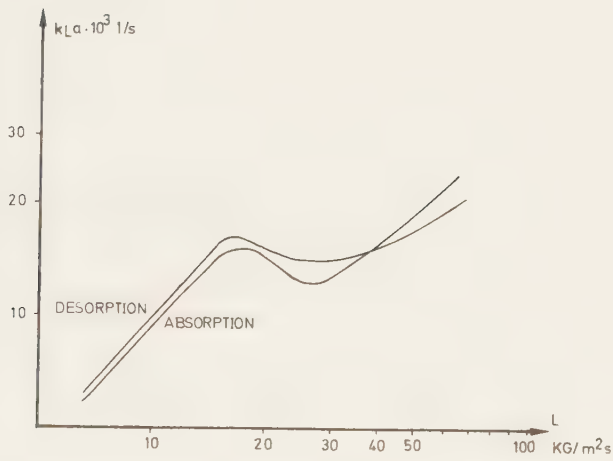


Figure 33. Effect of process, Plasdek at $z=3 \text{ m}$.

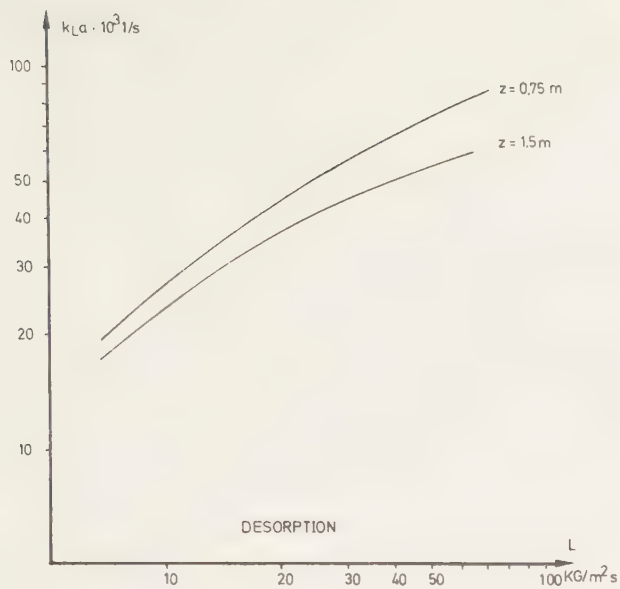


Figure 34. Effect of packing height, Intalox saddles.

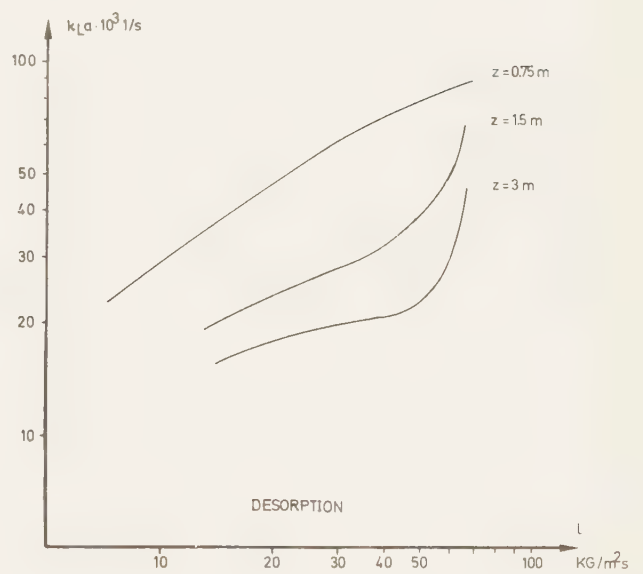
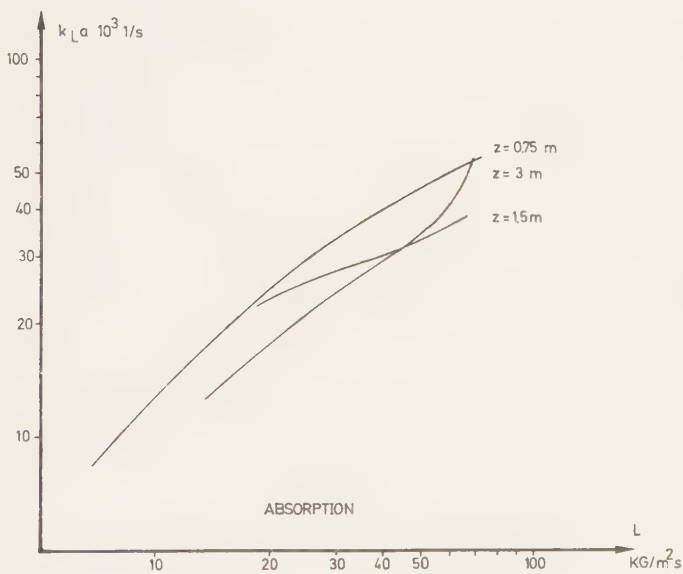


Figure 35. Effect of packing height, Tellerettes.

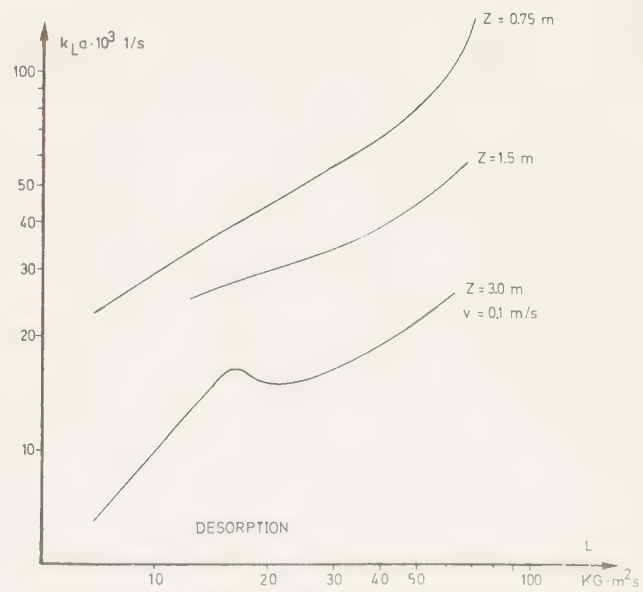
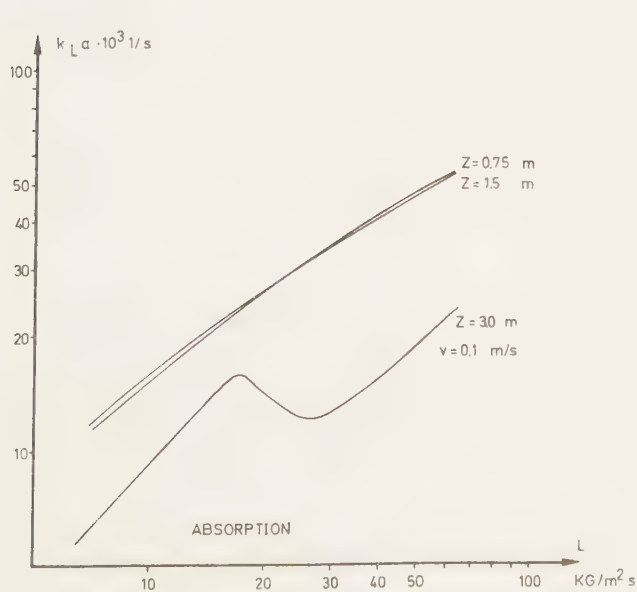


Figure 36. Effect of packing height, Plasdek.

Table 7

$$\gamma \text{ in } k_L a = \alpha(L)^\gamma \quad L < 15 \text{ kg/sm}^2$$

PACKING TYPE	Z m	ABSORPTION	DESORPTION
Plastic Intalox saddles	0.75	0.70	0.80
	1.5	0.41	0.77
Tellerettes	0.75	1.00	0.70
	1.5	1.00	0.67
	3	0.84	--
Plasdek	0.75	0.75	0.67
	1.5	0.80	0.67
	3	1.06	1.03

Table 8

Some liquid film data reported in literature
Packing size 25 mm (nom.)

Author	Ref No	Packing type 25 mm nominal size	System used	T °C	d m	z m
Yoshida, Miura	3	Porcelain Raschig rings	Abs CO ₂ -H ₂ O	12	0.121	0.76
Vivian, Whitney	6	Tile Raschig rings	Des O ₂ -H ₂ O	20	0.1, 0.36	0.6, 1.2
Vivian, Whitney	7	Ceramic Raschig rings	Des O ₂ -H ₂ O	20	0.203	0.63
Yoshida, Koyanagi	8	Ceramic Raschig rings	Abs CO ₂ -H ₂ O	20	0.12	0.41
		Ceramic Berl saddles	Abs CO ₂ -H ₂ O	20	0.12	0.41
Novella, Sancho	9	Intalox saddles	Abs CO ₂ -H ₂ O	20	0.3	0.4, 0.75
Sahay, Sharma	10	Ceramic Raschig rings	Abs CO ₂ -H ₂ O	0.2	0.87	
		PVC Raschig rings	Abs CO ₂ -H ₂ O	0.2	0.87	
		Ceramic Intalox saddles	Abs CO ₂ -H ₂ O	0.2	0.87	
		PP Intalox saddles	Abs CO ₂ -H ₂ O	0.2	0.87	
		SS Pall rings	Abs CO ₂ -H ₂ O	0.2	0.87	
		PP Pall rings	Abs CO ₂ -H ₂ O	0.2	0.87	
			Abs CO ₂ -H ₂ O	0.2	0.87	
Sherwood, Holloway	11	Ceramic Raschig rings	Des O ₂ -H ₂ O	25	0.50	1.24, 0.43
		Ceramic Berl saddles	Des O ₂ -H ₂ O	25	0.50	0.43
De Waal, Beek	13	Ceramic Raschig rings	Abs CO ₂ -H ₂ O	20	0.30	1.5
Shulman, de Gouff	14	Carbon Raschig rings	Des CO ₂ -H ₂ O	8	0.25	0.6
Mannford Doble	15	Ceramic Raschig rings	CO ₂ -H ₂ O			
		Ceramic Intalox saddles				
Merchuk et.al.	16	Carbon Raschig rings	Abs CO ₂ -H ₂ O	25	0.25	0.34

Table 9

m	Percent liquid wall flow of total flow		
	Plastic Intalox saddles	Tellerettes	Plasdek
0.75	15 - 20	3 - 10	15 - 20
1.5	15 - 35	4 - 15	20 - 30
3	20 - 40	8 - 27	14 - 30

9. CONCLUSIONS

The $k_L a$ -values presented in this paper are believed to be very valuable for designing packed towers. Three newly developed and highly efficient packings have been investigated. No $k_L a$ -values have yet been presented on the Tellerettes or the Plasdek. Likewise reported data on the plastic Intalox saddles are very limited. The packings in the present investigation have been tested under a wide range of operating conditions; the liquid range would presumably cover most industrial applications, the gas rate varies from well below loading up to flooding conditions, and the tower height has been increased four times. Furthermore the present set of data makes it possible to consider the effects of absorption versus desorption. The following conclusions may be drawn from results of the investigation.

1. The liquid side mass transfer data obtained for Plastic Intalox saddles, Tellerettes, and Plasdek are all of the same order of magnitude. A small superiority has been noticed for the Intalox saddles, especially when they are used for absorption purposes. This could be due to the fact that they were very densely packed.
2. The $k_L a$ -values obtained here are of the same order of magnitude as those obtained by other investigators. An examination of published data shows that when quantities such as temperature, packing height, column diameter, and packing density differ, the $k_L a$ -value will also differ significantly.
3. The $k_L a$ -values obtained in this investigation show roughly the same liquid flow dependency as do values from other investigations. When the upward gas velocity was much smaller than the downward liquid velocity, back-mixing of the gas was believed to occur. At these flow rates the gas velocity influenced the $k_L a$ -values.
4. At packing depths normally used in industry no effects from direction of mass transfer were noticed. However at more shallow packing depths higher packing efficiency was obtained when the gas was desorbed than when it was absorbed.
5. The height of the packed section seems to influence the mass transfer efficiency of the packings when they are used for desorption

purposes. Higher $k_L a$ -values were obtained at lower heights. The efficiency of the packings were affected to varying degrees; the efficiency of the Tellerettes, were greatly affected, the efficiency of the Plasdek somewhat less, and the efficiency of the plastic Intalox saddles only to a minor degree. However when the packings were used for absorption no height influence was noticed, provided that the packed section was no larger than about five times the tower diameter.

6. The Tellerettes showed poor self-distribution properties. When using the Tellerettes a good liquid distributor is essential.

ACKNOWLEDGEMENTS

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NOMENCLATURE

a	interfacial area per volume packing	$ \text{m}^2/\text{m}^3 $
c	liquid phase concentration	$ \text{kmole}/\text{m}^3 $
d	tower diameter	$ \text{m} $
e	difference in mass balance	$ - $
D_A	diffusion coefficient	$ \text{m}^2/\text{s} $
G	gas flow rate	$ \text{kg}/\text{m}^2 \cdot \text{s} $
He	Henry's law constant	$ \text{kmole}/\text{m}^3 \cdot \text{MPa} $
k_G	gas-film coefficient	$ \text{kmole}/\text{m}^2 \cdot \text{s} \cdot \text{MPa} $
k_g^a	gas-film coefficient	$ \text{kmole}/\text{m}^3 \cdot \text{s} \cdot \text{MPa} $
$k_{\text{H}_2\text{O}}$	forward rate constant; $\text{CO}_2\text{-H}_2\text{O}$	$ 1/\text{s} $
$k_{\text{H}_2\text{O}}^{-1}$	reverse rate constant; $\text{CO}_2\text{-H}_2\text{O}$	$ \text{m}^3/\text{kmole} \cdot \text{s} $
k_L	liquid-film coefficient	$ \text{m}/\text{s} $
k_L^a	liquid-film coefficient	$ 1/\text{s} $
K	equilibrium constant	$ \text{kmole}/\text{m}^3 $
K_w	dissociation constant of water	$ \text{kmole}/\text{m}^3 $
L	liquid flow rate	$ \text{kg}/\text{m}^2 \cdot \text{s} $
ℓ	fractional liquid hold up	$ - $
m	slope of equilibrium line = $\rho_m/\text{He} \cdot P$	$ - $
M	mole weight	$ \text{kg}/\text{kmole} $
Me-ions	metal-ion concentration	$ \text{kmole}/\text{m}^3 $
n	incremental step number n	
P_{CO_2}	partial pressure of CO_2	$ \text{MPa} $
$P_{\text{H}_2\text{O}}$	partial pressure of water	$ \text{MPa} $
P	barometric pressure	$ \text{MPa} $
$r(A^*, B^0)$	rate of reaction between dissolved A and the liquid component B	$ \text{kmole}/\text{m}^3 \cdot \text{s} $

R	rate of absorption	$ \text{kmole/m}^2 \text{ effective area} \cdot \text{s} $
s	standard deviation	$ - $
S	tower cross-sectional area	$ \text{m}^2 $
T	temperature	$ \text{C} $
v	superficial gas velocity	$ \text{m/s} $
x	mole fraction of CO_2 in liquid	$ - $
Vol_{CO_2}	volumetric content of CO_2 in gas	$ \% $
y	mole fraction of CO_2 in gas	$ - $
z	height of packing	$ \text{m} $
A	concentration of A = C	$ \text{kmole/m}^3 $
α	constant (eq. 24)	
ϵ	percent void	
γ	exponent (eq. 24)	
η	wetting rate	$ \text{m}^3/\text{m} \cdot \text{s} $
ρ	density	$ \text{kg/m}^3 $
ρ_m	mole density of liquid	$ \text{kmole/m}^3 $

Subscripts

1	tower end with highest concentration of CO_2
2	tower end with lowest concentration of CO_2
e	bulk equilibrium concentration
L	liquid
G	gas
*	equilibrium concentration
o	bulk concentration

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GAS-FILM MASS TRANSFER COEFFICIENTS FOR
THREE DIFFERENT TYPES OF PLASTIC PACKINGS

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Gas-film mass transfer coefficients have been determined for three types of plastic packings: Intalox saddles, Tellerettes and Plasdek. This was accomplished by absorption of SO_2 in NaOH-solutions. The operating conditions tested were primarily of interest in air pollution control.

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Abstract

Gas-film mass transfer coefficients have been determined by absorption of SO_2 in sodium hydroxide solutions for three plastic packings; Intalox saddles, Tellerettes, and Plasdek. A wide range of gas and liquid loads have been employed. The gas load was varied from 0.6 to $3.3 \text{ kg/m}^2 \cdot \text{s}$ and the liquid load from 2 to $50 \text{ kg/m}^2 \cdot \text{s}$. These wide ranges make the results especially valuable for designing air pollution control equipment.

The resulting $k_G a$ -values vary between $0.7\text{--}4 \text{ kmole/m}^3 \cdot \text{s} \cdot \text{MPa}$ depending on gas and liquid flow rates and type of packing. The Tellerettes have the superior mass transfer efficiency; 50 % better than the Intalox saddles at low gas flow rates. At higher flow rates the difference in $k_G a$ -values of the three packings becomes smaller, because of different gas and liquid flow rate dependencies. At comparable flow rates the Plasdek has the lowest $k_G a$ -value, while at the loading point, which is at higher flow rates for Plasdek, it has about twice as high $k_G a$ -values. It is also shown, for a gas-film controlled system, that a high gas load minimizes the tower volume, provided that $k_G a$ is independent of tower height. From experiments with deeper packing sections it is concluded that back-mixing of the gas-phase has no significant influence on the performance.

Table of Contents

	Page
1. Introduction	1.
2. Theory	2.
3. Experimental	7.
4. Results	10.
4.1 Effect of Gas Load	12.
4.2 Effect of Liquid Load	13.
4.3 Effect of Packing Height	13.
4.4 Comparing the three Packings	14.
4.5 Comparison with Literature Data	16.
5. Conclusions	19.
Acknowledgements	
Nomenclature	
References	

List of Figures

	Page
Figure 1. Experimental equipment	7.
Figure 2. $k_G a$ -values for plastic Intalox saddles	10.
Figure 3. $k_G a$ -values for Tellerettes	10.
Figure 4. $k_G a$ -values for Plasdek	11.
Figure 5. Comparison of $k_G a$ -values for the three packings	15.
Figure 6. Relative tower volume as a function of Gas Load	16.
Figure 7. Comparison with literature data: Intalox saddles	17.
Figure 8. Comparison with literature data: Tellerettes	17.
Figure 9. Comparison of literature data: Plasdek	18.
Figure 10. Literature data for different packings	19.

List of Tables

Table 1. Important experimental conditions used by some authors	3.
Table 2. Geometrical and packing characteristics of the packings	7.
Table 3. Exponents and constants of Eq. (5)	11.

1. INTRODUCTION

Gas absorption is a widely applied unit operation in industrial practice, both as an integrated part of a chemical process and as a tool for environmental protection. Many different types of apparatus, like packed column, tray columns, spray-scrubbers, impingement scrubbers etc. may be used. Although much work has been done, especially on gas absorption in packed columns, there is still a need of accurate and general design data. Performance data of many new packings and other mass transfer devices that can handle large gas quantities are limited. Such data are especially important in the field of air pollution control.

The present investigation provides gas-side mass transfer coefficients for three plastic packings suitable for air pollution control: Plasdek 12060, 25 mm nom. size Tellerettes, and 25 mm nom. size Plastic Intalox saddles. Together with pressure drop data, liquid-side coefficients, and the theory of mass transfer with reaction, a base for design of packed towers is established.

2. THEORY

When gas molecules are transported from a gas mixture to a liquid absorbent or vice versa, the resistance to the mass transfer is mainly confined to thin regions adjacent to the interface. This is the basis for the two-film model of mass transfer, i.e. all resistance is in a gas film and in a liquid film with negligible resistance at the interface. Quite often the resistance is concentrated in one of the two films, the mass transfer process is then termed either gas-film controlled or liquid-film controlled. Gas-film controlled systems are rather common in connection with air pollution control.

The resistance to mass transfer is expressed by the mass transfer coefficients. The magnitude of a mass transfer coefficient is dependent on both physical and chemical properties of the system and on the hydrodynamics and mass transfer area of the mass transfer device or packing used. Because both hydrodynamics and mass transfer area are different, the total efficiency may vary considerably between different packings or devices. The total mass transfer efficiency for a given equipment is established if the liquid- and gas-side coefficients are known, together with physical and chemical properties of the system. This is expressed by the $k_L a$ - and $k_G a$ -values as

$$\frac{1}{K_G a} = \frac{1}{k_G a} + \frac{1}{H \cdot k_L a \cdot E} \quad (1)$$

Here H is the solubility of the transferred gas in the pure solvent and E is the enhancement factor due to chemical reaction in the liquid. Expressions for the enhancement factor for different types of reaction may, for instance, be found in Danckwert's text book "Gas Liquid Reactions"¹.

The individual mass transfer coefficients, $k_L a$ and $k_G a$, are determined by performing experiments on mass transfer systems with the resistance entirely concentrated in one of the phases. Thus $k_L a$ -values are often established by absorption or desorption of CO_2 in water. Since CO_2 is sparingly soluble in water, the system is regarded as totally liquid-film controlled. Liquid-film mass transfer coefficients are rather common in literature. In an earlier work² $k_L a$ -values were presented for the same three packings as are examined here.

When establishing $k_G a$ -values varying techniques have been used. Table 1 summarizes the most important variables used by some authors.

Author	Ref. No	System used	Packing	Gas flow range kg/m ² s	Liquid flow range kg/m ² s	d _{tower} m	Z m	T °C	Nom. pack size
Fellinger	3	NH ₃ -water	Ceramic Raschig rings	0.27 - 1.35	3.67 - 6.1	0.46	0.22-0.66	Varying 5 - 20	10, 12.5, 25, 38, 50
		NH ₃ -H ₂ SO ₄	Ceramic Berl saddles	0.27 - 1.35	0.67 - 6.1				12.5, 25, 38
Akell and Talbott	4	NH ₃ -water	Fiberglass packing	0.27 - 2.03	2.7 - 28	0.4	1.8	10-15	a-590 m ⁻¹
Teller	5	NH ₃ -water	Tellerettes	0.4 - 1.0	5.8 - 3.4	0.15	1.2	-	19, 38
Meier, Stoecker	6	NH ₃ -water	Mellapak 250Y Pall rings	0.8 - 2.2	1	0.30	-	-	50
Manford Doble	7	NH ₃ -H ₂ SO ₄	Raschig rings	0.2 - 0.6	0.5 - 7	0.16	0.32	25	12.5, 16, 25
			Intalox saddles	0.2 - 0.6	2 - 7	0.15			
			Pall rings	0.2 - 0.6	0.7 - 7	0.23			
Sherwood and Holloway	8	NH ₃ -water	Carbon Raschig rings	0.05 - 1.4	0.8 - 2.4	0.25	0.4 - 0.8	25	25
		NH ₃ -H ₂ SO ₄	" "	0.12 - 0.5	2.2 - 2.5	0.25	0.4 - 0.8	25	25
		vap. of	Ceramic Raschig rings	0.03 - 2.5	3.2 - 4.3	0.50	0.2	25	38
Dwyer and Dodge	9	NH ₃ -H ₂ O	Carbon Raschig rings	0.1 - 1.3	0.2 - 1.3	0.30			12.5, 25, 38
Sahay and Sharma	11	Cl ₂ -NaOH	Ceramic Raschig rings						
			PVC-" "						
			Ceramic Intalox saddles	0.2 - 0.9	1 - 6	0.25	0.61	-	25
			pp-" "						
Johnstone and Singh	13	vap. of water NH ₃ -acetic acid SO ₂ -NaOH	SS Pall rings						
			pp Pall rings						
			Raschig rings	0.8 - 3.27	1.5		0.15		25
			Bregent coils	1.25 - 2.6	1.5		0.15		32
			Spiral tile	1.1 - 3.25	1.2	0.38, 0.13	0.78		75
			various conf. of woods and sheets	1.6 - 3.77	1.5				
Yoshida	16	vap. of water	Stoneware	0.04 - 1.4	0.7 - 8	0.10	0.13, 0.19		25 Three carrier gases
Yoshida and Koyanagi	17	Methanol-water vap. of water	Raschig rings	0.1 - 1.4	0.28 - 4 1.25 - 7	0.12	0.1, 0.2		15, 25
McAdams, Pohlentz and St John	18	vap. of water	Carbon Raschig rings	0.5 - 1.4	0.7 - 7	0.15	0.15-0.30		25
Shulman, Savini and Edwin	19	Methanol-water vap. of water	Porcelain Berl saddles Porcelain Raschig rings	0.2 - 1.7	2 - 7	0.30	0.75	-	12.5, 25, 38

Table 1. Important experimental conditions used by some authors.

Fellinger's³ work on absorption of ammonia in water is often referred to for $k_G a$ -values. Many other investigators have also used this system⁴⁻⁸. As Dwyer and Dodge⁹ showed in 1941 the ammonia-water system could not be regarded as entirely gas-film controlled under any of their experimental

conditions. The gas-film resistance varied between 40 and 90 percent of the total resistance. The liquid-film resistance could be further reduced by reacting ammonia with an acid, and thus increasing the enhancement factor. Fellingner³, Sherwood-Holloway⁸, and Manford Doble⁷ have absorbed ammonia in sulphuric acid at different strengths, resulting in greater $K_G a$ -values. Theoretically, the $K_G a$ -value increases with increasing acid strength until all absorbed gas molecules react instantaneously at the interface. No concentration gradient is then present in the liquid-film and all of the resistance is in the gas-film. This has been confirmed in an earlier study of ammonia absorption in phosphoric acid¹⁰.

The criterion for the system to be fully gas-film controlled can be formulated as:

$$k_G a \cdot y_a \cdot P < k_L a \cdot B^0 \cdot D_B / q \cdot D_A \quad (2)$$

This criterion implies that the concentration of the soluble gas is zero at the interface between gas and liquid everywhere in the tower due to an instantaneous and irreversible reaction between A and B. Such a gas-liquid system is hypothetical, but many systems approximately fulfil the conditions at low concentration of the soluble component in the gas phase and high concentration of the reactant in the liquid. Ammonia absorption in various acids is an example. Also absorption of an acidic gas in an alkaline solution has been employed. In addition to his study of ammonia absorption in sulphuric acid, Manford Doble⁷ also absorbed sulphur dioxide and hydrogen chloride in caustic soda solution, both systems being fully gas-film controlled. Sahay and Sharma¹¹ used the system Cl_2 -NaOH when they examined several 0.025 m plastic packings. The system SO_2 -NaOH has been used by Shende and Sharma¹² in a study of concurrent absorption and by Johnstone and Singh¹³ in an early investigation of packings suitable for removing SO_2 from waste gases. In a recent investigation of the Sulzer packing, Bomio¹⁴ absorbed both SO_2 and NH_3 under conditions that ensured full gas-phase control. Other gas-liquid systems, like absorption of various amines in acids and HCl or methanol in water can be made to fulfil the requirements. Mehta and Sharma¹⁵ have used a variety of different systems when investigating the effect of diffusivity on $k_G a$.

Mehta and Sharma, in addition to absorption, also vaporized various liquids. In this case the liquid phase only consists of the pure solute and thus does not exhibit any concentration gradient. Under adiabatic conditions the only gradient is in the gas film and the system is fully gas-film controlled. Several authors have used evaporation of water into different carrier gases to establish gas-film coefficients^{8,16,17,18}. However, the mass transfer coefficients for vaporization are, in general, significantly greater than those for absorption under total gas-film control. Shulman and coworkers¹⁹ have found that this is mainly due to different effective interfacial areas for different types of operation; stagnant portions of the liquid within the packing are effective for vaporization but not for absorption. By making experiments in a short wetted wall column with a constant active interfacial area Vivian and Schoenberg²⁰ supported this theory. They report equal true mass transfer coefficients both for vaporization of water, desorption of methanol, and absorption of ammonia, when correction is made for different diffusivities.

In this work $k_G a$ -values have been evaluated by absorption of sulphur dioxide in caustic soda solutions at a high pH to ensure full gas-film control. Liquid and gas flow rates have been selected mainly in a range suitable for air pollution control. Packed sections of different height have been investigated to determine height effects on the performance of the packings.

When SO_2 is absorbed in sufficiently strong aqueous sodium hydroxide solutions it reacts instantaneously and irreversibly according to the following overall reaction:



The solution must be strong enough to keep the reaction plane at the interface. As long as condition (2) is fulfilled it is considered to be so. For typical operating conditions this means that the pH must be greater than 12.3 at the bottom of the column. When the reaction takes place at the interface, the interfacial partial pressure of SO_2 , y^* , will be zero and $k_G a$ -values can easily be evaluated from

$$k_G a = \frac{G}{M_G \cdot Z \cdot P} \cdot \ln \frac{y_1}{y_2} \quad (4)$$

where y_1 and y_2 are the mole fraction at bottom and top of the column, respectively; G the gas load, M_G gas molecular weight, Z tower height, and P total pressure. This equation is easily derived from the two film model by setting $y^* = 0$.

3. EXPERIMENTAL

The experimental equipment is schematically outlined in figure 1.

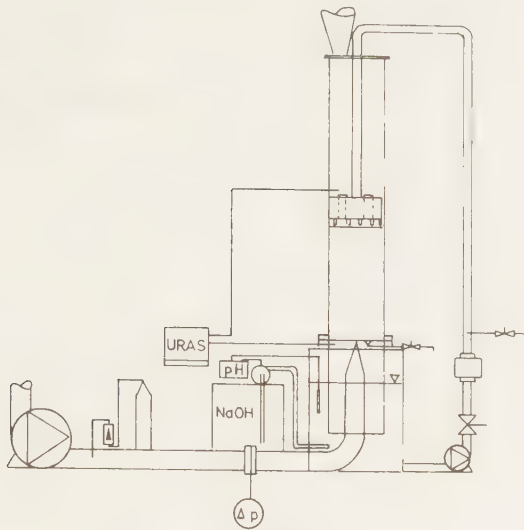


Figure 1. Experimental equipment

It mainly consisted of a tower made of glass fibre lined polyester having an inside diameter of 0.3 m. The tower was packed to different heights: 0.375 m, 0.75 m, and with the Plasdek also to 1.5, and 2.4 meters. The Tellerettes and the Intalox saddles were packed dry in a random fashion. The Plasdek was packed to the desired height in three sections at right angles to each other; two blocks of forty percent of the height each and one of twenty percent. No blocks were higher than 0.3 m. When deeper beds were required the number of blocks were increased. The blocks contained no drainage tips. Some important geometrical data and packing characteristics are gathered in table 2.

Geometrical and packing characteristics of the packings				
Type	nominal size m	a m^2/m^3	ϵ %	Number of packing pieces per m^3
Plastic Intalox saddles	0.025	210	91	60 000
Tellerettes	0.025	180	87	40 000
Plasdek 12060	-	220	97	-

Table 2. Geometrical and packing characteristics of the packings

The liquid was carefully distributed over the packing and continuously

circulated, at a controlled temperature of about 18°C. The pH was maintained at a predetermined high value by a standard pH-control system. Air at atmospheric pressure was made up with SO₂ to a concentration of about 1000 ppm and blown by a fan through the tower. The flow rate was measured by interchangeable orifices. Sampling points for gas and liquid were placed immediately below and above the packed section in order to minimize end effects. All liquid samples were analyzed on hydroxide content by titration, in order to be certain that the experimental conditions fulfilled the requirements for full gas-phase control. The gas concentration was determined by an infrared gas analyzer. This was carefully calibrated with test gas at the maximum sensitivity range of the instrument. Thus, the leaving gas concentration could be analyzed down to ppm-level. The temperature of the leaving gas was lower than the surroundings, which ensured that no vapour condensed at the tube wall. The entering gas could be analyzed in two different ways; by IR or by measuring the amount of SO₂ added, with a rotameter. The IR-analysis occasionally could be quite unreliable, because liquid could be entrained in the sample lines at higher liquid flow rates. The rotameter readings suffered from end effects, which could not be avoided with the particular design of the gas injector. By measuring the absorption below the support plate, without any packing, a correction factor was introduced for the end effects. If the gas concentration, as measured with the rotameter, was corrected with this factor the true ingoing gas concentration to the packed section was established. At lower liquid flow rates a good consistency was shown with the IR-analyses.

Each experiment was performed by first adjusting liquid and gas flow rates. After the liquid had attained the desired temperature of 18°C, sulphur dioxide was added to the air stream. Enough SO₂ was added to keep a concentration of about 1000 ppm when the gas entered the packing. After about twenty minutes equilibrium was well established and concentration measurements were made. A whole test series, i.e. one packing at one height, were performed with the same liquid circulating. Only NaOH was added to keep the pH constant. Generally, an increasing ionic strength is considered to have only minor influence on the performance of a packing. Measurements of density and viscosity of the liquid showed that the density varied about 5 percent and viscosity about 50 percent from the first experiment in the series to the last. These variations in physical properties probably have no influence on the hydrodynamics because of the turbulent character of flow in packed columns.

When all of the concentrations of an experiment were established, the k_G a-value could easily be evaluated from equation (4). The k_G a-values thus determined will have a maximum error of about ± 16 percent according to standard estimation procedure. An average error of about ± 10 percent may also be established from the standard deviation of the measurements.

4. RESULTS

The resulting mass transfer coefficients for the three packings are displayed in figures 2-4. In the three figures $k_G a$ -values are plotted as a function of liquid load with gas load as parameter. The figures present $k_G a$ -values for a very wide range of operating conditions; from below loading to above flooding at liquid loads below minimal wetting rate to extremely high ones. For air pollution control data at higher gas flow rates and lower liquid flow rates are especially valuable.

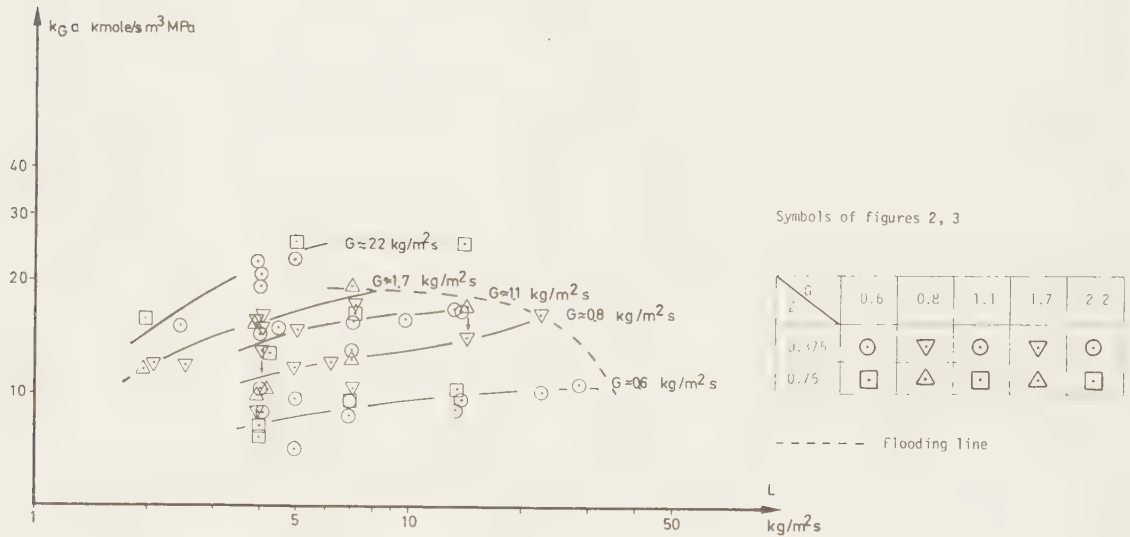


Figure 2. $k_G a$ -values for plastic Intalox saddles

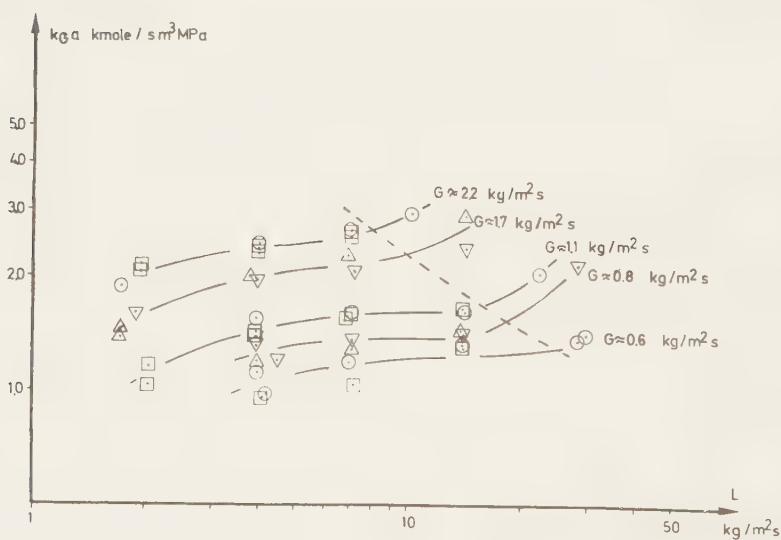


Figure 3. $k_G a$ -values for Tellerettes

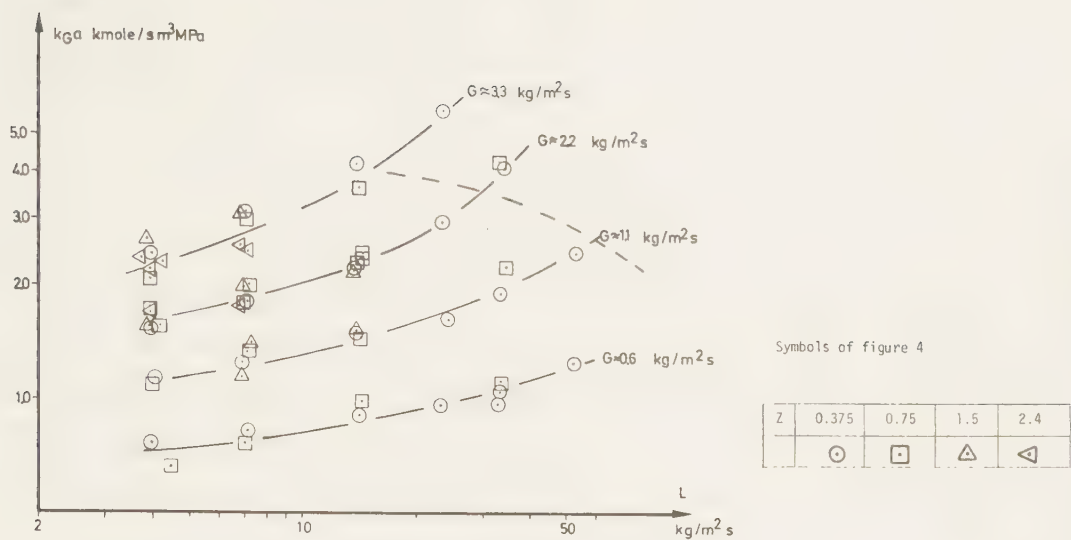


Figure 4. k_Ga -values for Plasdek

When the k_Ga -values obtained at operating conditions below minimum wetting and above incipient flooding are omitted, the remaining values can be correlated by a multiple regression analysis as

$$k_Ga = c \cdot G^n \cdot L^m \quad (5)$$

Table 3 lists the constants and exponents for the various packings employed. In the table are also shown the standard deviation together with maximum and mean deviation of predicted values from experimental values.

$$k_Ga = a \cdot G^n \cdot L^m$$

Packing	a	n	m	Deviation, %		s
				Min	Max	
Plastic Intalox saddles	0.92	0.69	0.19	5.0	24	0.146
Tellerettes	1.17	0.65	0.14	5.2	13	0.101
Plasdek	0.73	0.67	0.23	5.5	15	0.096

Table 3. Exponents and constants of Eq. (5)

The correlation formula is not dimensionless and should be used with SI-units. The formula merely shows the mean influence of gas and liquid flow rates for the gas-film controlled system SO_2 -air-NaOH solution at atmospheric pressure. When used for other systems proper correction for other variables like diffusivity, viscosity, density, and pressure should be made.

4.1 Effect of Gas Load

The mass transfer efficiency of a packing for a gas-film controlled system is greatly dependent on gas load. Generally an exponent of 0.7 is anticipated. About the same exponent is found in this investigation. Although a regression analysis with the gas flow as the only independent variable would give somewhat varying exponents of G at different liquid loads, there is a general trend of lower gas flow dependency for the Tellerettes. From a theoretical viewpoint this might also be expected. It was noticed by visual observations, that the liquid distributed in droplets in a bed of Tellerettes, while the Intalox saddles and the Plasdek distributed the liquid in a film. Literature correlations of gas phase mass transfer data for simple and idealized flow conditions suggest that there is a higher gas flow dependency for mass transfer to a film than to a drop.

4.2 Effect of Liquid Load

Two major effects of an increasing liquid flow rate can be anticipated; an increase in available mass transfer area and a higher relative velocity between gas and liquid phases. Sahay and Sharma have shown that at liquid loads up to $6 \text{ kg/m}^2 \cdot \text{s}$ it is the mass transfer area, a , that increases while the true gas-film coefficient, k_G is independent of liquid flow rate. Most literature gas-side mass transfer data have been determined at low liquid flow rates, as is evident from both table 1 and figure 9. Normally it is anticipated that $k_G a$ varies as $L^{0.4}$ in this flow region. From figures 2 and 3 it is seen that when L is below $4 \text{ kg/m}^2 \cdot \text{s}$ the slope of 0.4 is established, but above this flow rate the slope has only about half of this value. At a too low liquid flow rate the available packing surface may be badly used because of poor wetting. According to the criterion for wetting proposed by Morris and Jackson²¹ most published $k_G a$ -values have been determined at a liquid flow below the minimum wetting rate. (For packings with $a \approx 200 \text{ m}^{-1}$ this means $L_{\min} \approx 4 \text{ kg/m}^2 \cdot \text{s}$.) Originally this minimum wetting rate was established for water on ceramic packings. Naturally, materials and substances with different surface tensions exhibit different wetting. In the simple expression of Morris and Jackson no account is taken of this. On the other hand, the effect of different surface tensions may for instance be outweighed by impurities on the surface, chemical interaction between the liquid and the surface, and capillary forces. For water sol-

utions of NaOH on polypropene plastic packings this seems to be so. Although pure plastic surfaces are less wettable than ceramic surfaces and NaOH-solutions have higher surface tensions than pure water, there seems to be less wetting below a minimum wetting rate of $4 \text{ kg/m}^2 \cdot \text{s}$ as proposed by Morris and Jackson.

The relationship between the $k_G a$ -values and L found in this investigation is considerably lower than normally used in literature. The reason for this is the wide flow regime investigated. At the high liquid flow rates encountered here the interfacial area will not increase as fast as before; not only the wetted area increases but also the thickness of the film. At some higher flow rate all available packing surface is used for mass transfer, and the $k_G a$ -value only increases as a result of increasing relative velocity between the phases.

As previously mentioned the packings were investigated over a wide range of gas and liquid flows. Some points in figure 2, 3, and 4 were established at or above flooding. The approximate location of the flooding points, as obtained from pressure drop measurements, are plotted in the figures. Above this flooding line the $k_G a$ -values increase considerably, which is quite logical. Such flow conditions are however rarely used in packed columns, because of unstable performance and liquid entrainment. Normally packed towers are operated in the loading to flooding regime.

Between the minimum wetting rate and location of flooding the $k_G a$ -values for the Tellerettes show surprisingly low liquid flow dependency. A plausible explanation for this behaviour may be that the number of droplets formed in the packing is almost constant with increasing diameters as the liquid flow increases. If the droplets are regarded hypothetically as rigid spheres, the correlation of Ranz and Marshall²² shows that the true gas-film coefficient, k_G , decreases with increasing drop diameter. In addition, the increase in surface area is less than if the number of droplets were increased. Thus, for the product of the two, $k_G a$ is relatively unaffected by the liquid flow rate.

4.3 Effect of Packing Height

The great absorption efficiency of SO_2 in hydroxide solution made it necessary to establish the $k_G a$ -values at a low packing height. With greater heights the leaving gas stream could be very poor in SO_2 . The measurement of a low

concentration, below a few ppm, was rather unreliable with the disposable analyzer. The two heights of 0.375 m and 0.75 m both resulted in gas concentrations in a suitable range for analysis. However, end-effects could seriously influence the data, turning up in different $k_G a$ -values at different heights. As is apparent from figure 2-4 this is not so. This indicates that the correction used for the entering gas takes account of all important end-effects sufficiently well.

When designing a packed tower for air pollution control a higher tower is generally necessary because of rigorous legal requirements on the effluent. Likewise a less efficient absorbent must often be chosen, because of practical limitations in treatment of the waste liquor or cost of chemicals. Furthermore, generally large gas streams have to be handled, which requires high gas loads to minimize the capital investment costs.

At a fixed ratio between gas and liquid loads the tower height will increase as $G^{1-(m+n)}$ as is evident from equation (4) and (5). However maldistribution of liquid and back-mixing may reduce the mass transfer efficiency when the tower height increases.

An investigation of the influence of tower height was performed by testing the Plasdek packing at two additional heights: 1.5 and 2.4 meters. When using this packing a high tower may often be advisable because of a smaller mass transfer efficiency and a higher gas capacity. Likewise the open configuration makes the packing sensitive to back-mixing, which has been demonstrated at low gas flow rates^{2,3}. The resulting $k_G a$ -values are plotted in figure 4. As can be seen, no systematic difference in the mass transfer efficiency at the different heights exists. At some further increased tower height maldistribution of the liquid will presumably occur. By installation of wall-wipers and redistributors this is relatively easy to cope with.

4.4 Comparing the Three Packings

Figure 5 compares the mass transfer efficiency of the three packings at three different gas loads. The Tellerettes have the superior mass transfer efficiency. A fifty percent difference is noticed at most. The difference diminishes when the gas and liquid load increases as also

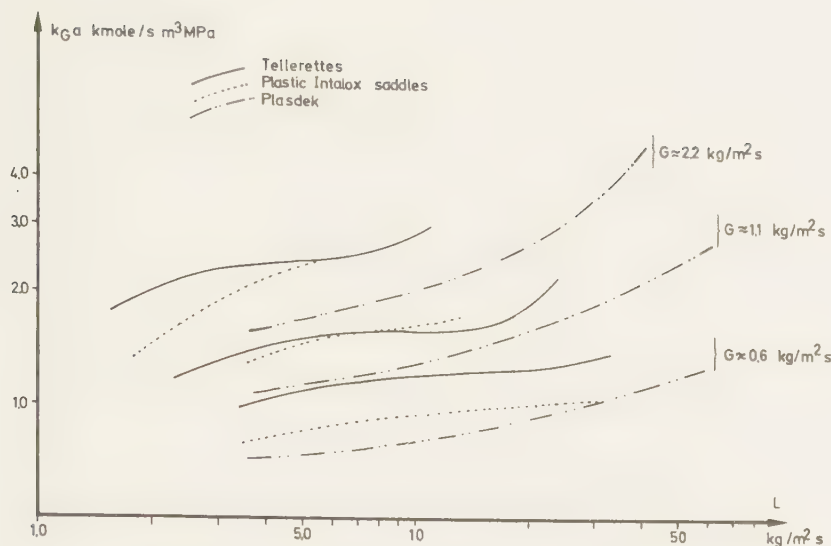


Figure 5. Comparison of k_Ga -values for the three packings

is apparent from equation (5). When comparing the packings it should also be borne in mind that the pressure drop is quite different. The Plasdek packing has a much lower pressure drop. The operating regime is thus extended to larger gas loads, which is of great importance in connection with air pollution control.

It is also interesting to examine what tower volume is needed to absorb a certain amount at different gas loads and constant gas to liquid flow ratio. Since the tower volume, V , is the product between the cross section and tower height it is easily shown from equations (4) and (5) that

$$\frac{V_1}{V_2} = \frac{C_2}{C_1} \cdot \left(\frac{G_2}{G_1}\right)^{m+n} \quad (6)$$

By taking the volume required when using the Plasdek at $G = 1.1 \text{ kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ as a basis, the relative volumes required at different gas loads and packings are established as plotted in figure 6. From the figure it is obvious that smaller towers are required at higher gas loads, saving capital investment costs at the expense of increasing pressure drop. The Plasdek with its lower pressure drop should preferably be operated at a higher gas load and may thus be very competitive when larger gas volumes are to be handled. It should be pointed out that figure 6 is only valid when equation (5) is valid. Gas loads must not be chosen, at a predetermined

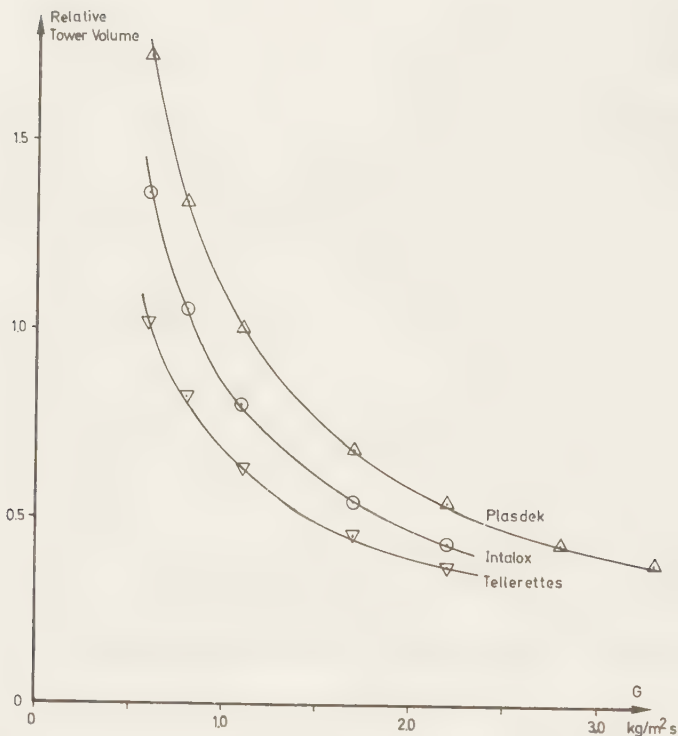


Figure 6. Relative tower volume as a function of Gas Load

gas to liquid ratio, that exceed the flooding point. Likewise the liquid flow rate may not be below the minimum wetting rate.

4.5 Comparison with Literature Data

In figures 7, 8, and 9 a comparison is made with other gas-side mass transfer data reported for the packings. In addition, figure 10 presents data for some other packings reported in the literature. All mass transfer coefficients have been corrected to the SO_2 -system by assuming $k_G a$ to be proportional to $\sqrt{D}^{15}$. Generally gas-phase mass transfer data are rather scarce in the literature at the high gas and liquid load employed here. Manford Doble⁷ absorbed ammonia in sulphuric acid solution. A rather good agreement is established with the present data as can be seen in figure 7. Data of Sahay and Sharma¹¹, as obtained with the system $\text{Cl}_2/\text{air-NAOH}$ deviate a good deal. The end-effects, however, were not specially corrected for by Sahay and Sharma. They performed some tests at two heights and concluded that there were no appreciable end-effects, which seems to be rather unrealistic with their experimental set up and method of analysis. The data in figure 8 for the Tellerettes were originally obtained by Teller by absorption of NH_3 in water. Some liquid-film resistance may

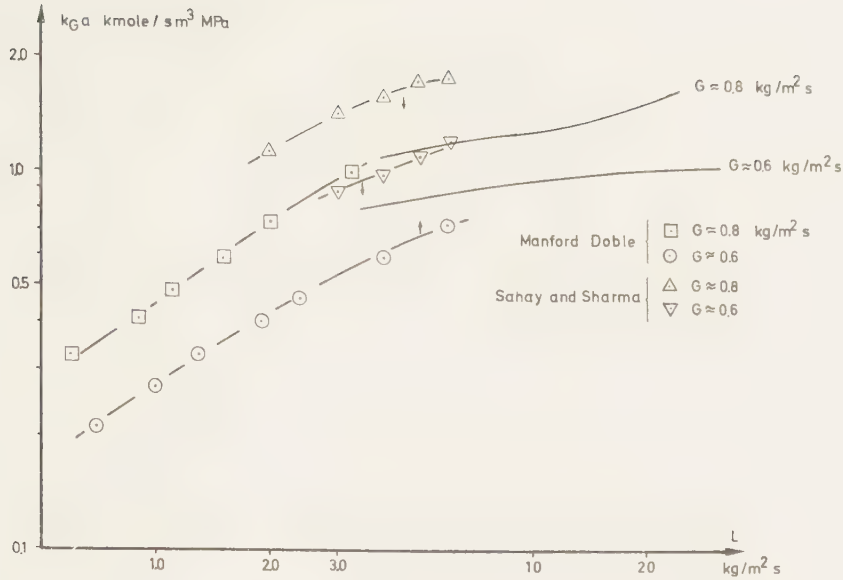


Figure 7. Comparison with literature data: Intalox saddles

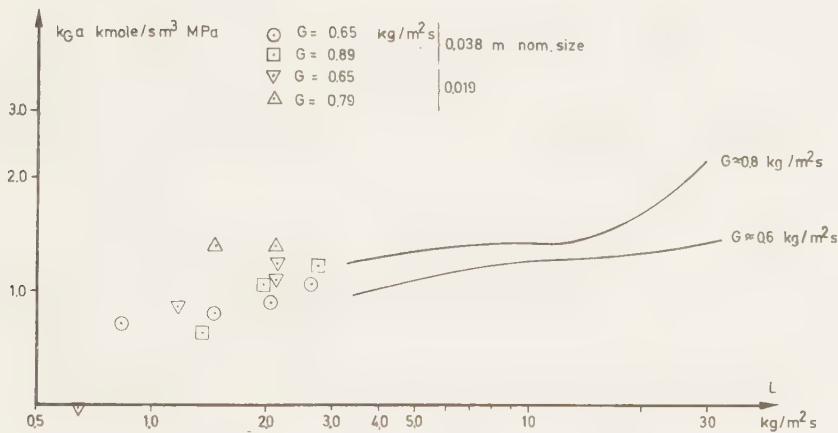


Figure 8. Comparison with literature data: Tellerettes

thus be included in the data, especially at the low liquid flow rates used. On the other hand no correction was made for end-effects.

Figure 9 compares data evaluated with three different systems for the Plasdek. The air-water data were obtained as heat transfer coefficients from cooling tower design manuals. By using the Chilton-Colburn analogy between heat and mass transfer with the Lewis number equal to one, mass transfer coefficients were easily calculated. However, as mentioned earlier, gas-film coefficients are only obtained under adiabatic conditions for the air-water system. The prevailing data thus incorporates some liquid-film resistance. Both the NH_3 -data and air-water data were

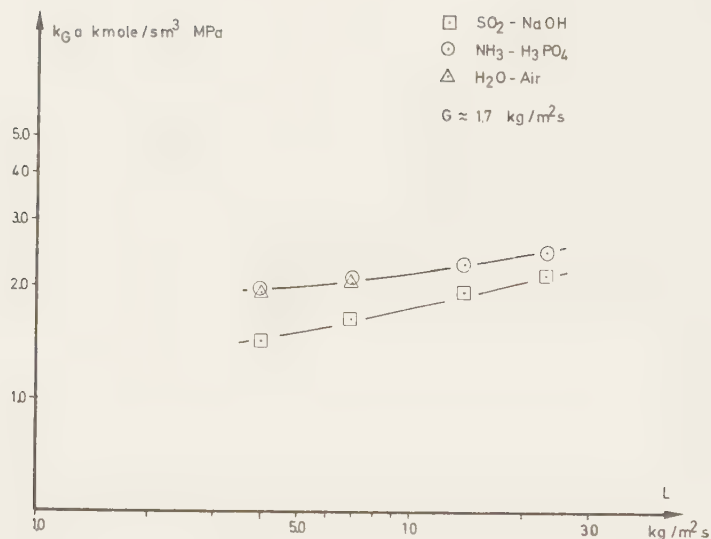


Figure 9. Comparison of literature data: Plasdek

not corrected for end-effects which is probably the main reason for the higher mass transfer efficiency. Under the experimental conditions used for the NH_3 -acid system¹⁰ it was impossible to establish the end-effects. The $k_G a$ -values will be about 15 percent lower if the same correction factors are used for the NH_3 runs as were used for the SO_2 runs.

Figure 10 shows good consistency between the present data, data from Manford Doble⁷ and data from Yoshida and Koyanagi¹⁷. Data reported by Sahay and Sharma¹¹ deviates a good deal, probably because of end effects, as previously mentioned. The mass transfer efficiency for the Sulzer packing seems to be superior to the others'. However Bomio¹⁴ does not mention anything about end effects, the geometrical area of the Sulzer BX is about twice as large and the gas load employed was somewhat higher. The Pall rings also seem to be an efficient packing in the flow range investigated. It is evident, however, from the mass transfer efficiencies presented, that gas and liquid loads play an important role; one packing may have the superior $k_G a$ at a particular set of flows but another packing in some different flow range. When making the final choice between packings care should also be taken about price and pressure drop.

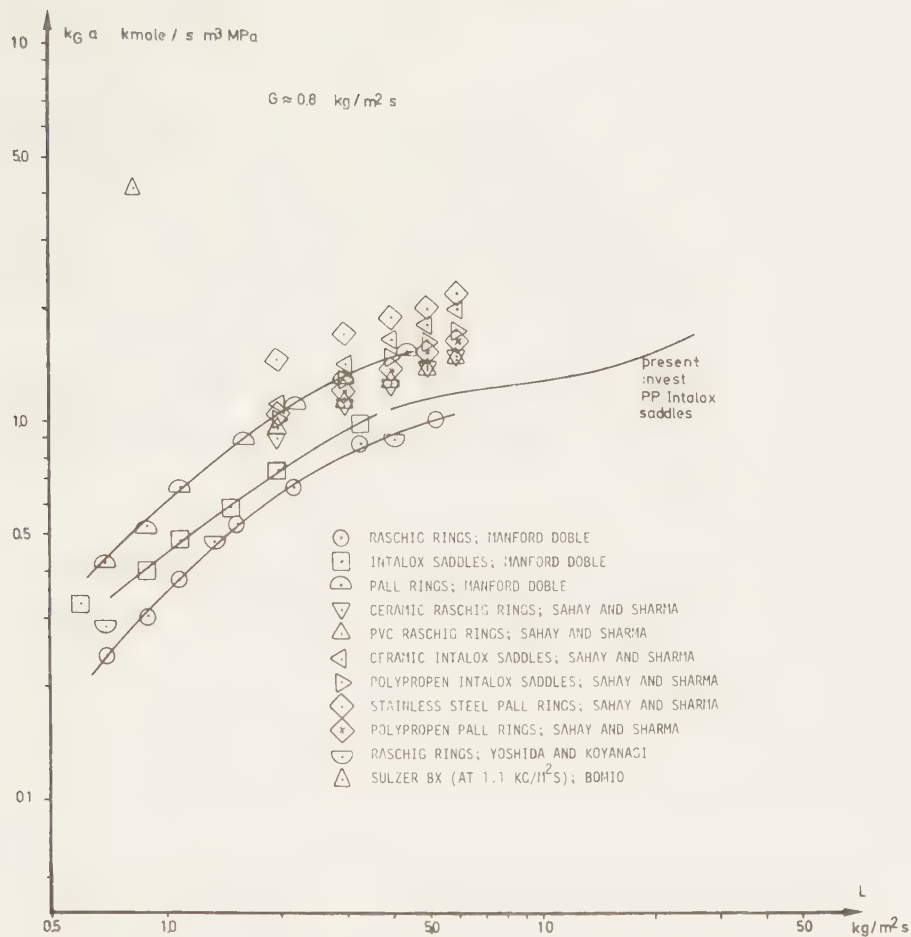


Figure 10. Literature data for different packings

6. CONCLUSIONS

The gas-side mass transfer coefficients presented for the three packings, Intalox Saddles, Tellerettes, and Plasdek, are believed to be very valuable in designing packed towers. A wide range of operating conditions, not previously reported in literature, but frequently used in industrial practice, has been investigated. The following conclusions may be drawn from the present set of data.

1. The $k_G a$ -values of the three packings differ a great deal. The Tellerettes have the superior efficiency, at most 50 % greater $k_G a$ than the others.
2. Gas and liquid loads affects the mass transfer of the packings differently. At higher gas load the efficiency of the Intalox saddles and the Tellerettes becomes equal. The Plasdek is more sensitive to liquid load.
3. Lower liquid load dependency is established in this paper than usually reported in literature. This is because much larger liquid flow rates have been investigated. Above the minimum wetting rate an exponent on L of about 0.2 seems relevant.

4. No effects of back-mixing have been noticed. This is especially interesting in air pollution control where high towers are needed.
5. To minimize the tower volume the packings should preferably be used at a high gas load.
6. The Plasdek may be used at higher gas loads than the others without flooding. The Plasdek may be compared with packings of larger nominal size.
7. A rather good consistency is found with literature data for different gas-phase controlled systems. However care must be taken of end effects.

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Nomenclature

a	Specific interfacial area	$\text{m}^2 \cdot \text{m}^{-3}$
B^0	Concentration of reactants	$\text{kmole} \cdot \text{m}^{-3}$
C	Constant of Eq. (5)	-
D	Diffusion coefficient	$\text{m}^2 \cdot \text{s}^{-1}$
d	Tower diameter	m
E	Enhancement factor due to reaction	-
ϵ	Percent void	%
G	Gas flow rate	$\text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$
H	Gas solubility	$\text{kmole} \cdot \text{m}^{-3} \cdot \text{MPa}^{-1}$
K_G^a	Overall mass transfer coefficient	$\text{kmole} \cdot \text{m}^{-3} \cdot \text{s}^{-1} \cdot \text{MPa}^{-1}$
k_G^a	Gas-side mass transfer coefficient	$\text{kmole} \cdot \text{m}^{-3} \cdot \text{s}^{-1} \cdot \text{MPa}^{-1}$
k_L^a	Liquid-side mass transfer coefficient	s^{-1}
L	Liquid flow rate	$\text{kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$
M_G	Molecular weight of the gas phase	$\text{kg} \cdot \text{kmole}^{-1}$
m	Exponent of L in Eq. (5)	
h	Exponent of G in Eq. (5)	
P	Total Pressure	MPa
q	Stoichiometric coefficient in reaction of A and B	-
S	Tower cross section	m^2
s	Standard deviation	-
T	Temperature	$^{\circ}\text{C}$
y	Mole-fraction absorbable gas component in gas phase	-
Z	Tower height	m

Subscripts

A	Absorbable gas component
B	Reactant
G	Gas
1	Conditions at the bottom
2	Conditions at the top

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PRESSURE DROP AND FLOODING PROPERTIES
FOR THREE DIFFERENT TYPES OF PLASTIC
PACKINGS

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Pressure drop and flooding properties have been measured for countercurrent air and water flow in beds of three types of plastic packings: Intalox saddles, Tellerettes, and Plasdek. These measured values have been correlated with pressure drop and flooding point correlations presented in the literature.

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Abstract

Pressure drops and flooding points have been determined for three different plastic packings: Intalox saddles, Tellerettes, and Plasdek. The liquid and gas flow rates have been varied over a wide range of operating conditions. The result shows that the Plasdek has a very low pressure drop compared with the Intalox saddles and Tellerettes. Likewise, the Plasdek permits higher gas load at every liquid load before flooding.

The measured pressure drops have been correlated with a simple model based upon the interaction of liquid holdup on dry pressure drop. With this model it was possible to correlate the wet pressure drop within 20 percent. The generalized pressure drop correlation could not be used. The flooding points were best correlated by the introduction of a "flooding" factor in the correlation originally proposed by Sherwood et. al.¹⁰.

List of Contents

	page
1. INTRODUCTION	1.
2. THEORY	2.
2.1 Dry Pressure Drop	3.
2.2 Liquid Holdup	4.
2.3 Wet Pressure Drop	4.
2.4 Flooding	6.
3. EXPERIMENTAL	10.
4. RESULTS	12.
4.1 Prediction of Pressure Drop in Packed Columns	17.
4.2 Prediction of Flooding in Packed Columns	22.
5. CONCLUSIONS	28.
Acknowledgements	
Nomenclature	
References	

List of Figures

	Page
Figure 1. Modified generalized pressure drop correlation	6.
Figure 2. Pressure drop - Intalox saddles	12.
Figure 3. Pressure drop - Tellerettes	13.
Figure 4. Pressure drop - Plasdek	13.
Figure 5. Liquid holdup	16.
Figure 6. Dry pressure drop correlation - Bemmer and Kalis ⁴	18.
Figure 7. Dry pressure drop correlation - Ergun ¹	19.
Figure 8. Correlation of liquid holdup	20.
Figure 9. Flooding point correlation - Perry ⁵	23.
Figure 10. Flooding point correlation - Zhavoronkov ¹⁸	24.
Figure 11. Flooding point correlation - Mersmann ¹⁹ , Reichelt ¹⁶	24.
Figure 12. Flooding point correlation - Eduljee ²¹	25.
Figure 13. Flooding point correlation with flooding factors	26.

List of Tables

Table 1. Packing characteristics	10.
Table 2. Slope of the dry pressure drop line	14.
Table 3. Packing factors	16.
Table 4. Correlation of Λ -values	20.
Table 5. Correlation of liquid holdup	20.
Table 6a. Constriction factors for wet pressure drop	21.
Table 6b. Constriction factors for wet pressure drop	22.
Table 7. Packing factors - method of Eduljee ²¹	25.
Table 8. Flooding factors	26.

1. INTRODUCTION

The gas handling capacity of a packed tower is determined by the pressure drop imposed by the two phase flow of gas and liquid. When the tower is operated countercurrently, an upper limit is fixed for the gas handling capacity at every liquid flow rate by the flooding point. At this particular gas and liquid load combination the liquid cannot flow downward over the packing due to interfacial stresses imposing standing waves. Although many attempts to predict the flooding point have appeared in the literature, there is still a need for further work. This is especially true of modern packings with high efficiency and capacity. In this report three different plastic packings: Intalox saddles, Tellerettes, and Plasdek, have been investigated. Experimentally determined countercurrent pressure drops and flooding points have been compared with predictions from a variety of models presented in the literature.

2. THEORY

The fluid flow in a packed bed is generally considered as a flow in a collection of convoluted channels formed by the packing bodies. Basically, models are based upon three types of channels: straight channels with a length equal to the height of the bed, winding channels, and channels with constrictions. By using these models there have been some successes in predicting liquid holdup, as well as dry and wet pressure drop below loading conditions. With a model of flow past immersed bodies the success has been limited and this model is rarely used.

For the flooding phenomenon, no theoretical models based upon the formation of standing waves have been proposed. Empirical correlations based upon theoretical and dimensional considerations have been developed instead. However, these models are mostly concerned with packings of ring or saddle type. It is still doubtful whether the models also are valid for the Tellerettes, where the liquid flows in droplets, or for the Plasdek packing with its open configuration.

When the logarithm of the pressure drop in a packed column is plotted against the logarithm of the gas velocity at different liquid loads, essentially straight lines are obtained. At zero liquid flow the slope is about 1.8-2.0. When liquid flow is imposed countercurrently the void decreases and the gas velocity increases. In addition there is an increase in relative velocity between the phases and eventually also an increase in roughness of the interface, both factors resulting in larger interfacial stresses. With increasing gas loads, different hydrodynamic regimes may be observed. With film flow, at comparatively low gas and liquid loads, the liquid holdup is independent of gas load. The pressure drop curves are parallel to the curve for the dry packing but above it because of the smaller void fraction. As the gas load increases the loading point is reached, where there is a strong gas load influence on liquid holdup. At even higher gas loads the packing begins to flood and the pressure drop curves are essentially vertical. At this point so much liquid is accumulated in the packing that a phase inversion occurs with the liquid becoming the continuous one with the gas phase dispersed in it. Normally, packed towers are operated close to loading conditions. However, this condition may be hard to establish in a pressure drop chart. The flooding point is used instead, followed by a 30-50 percent reduction of the gas velocity to obtain the operating condition.

2.1 Dry Pressure Drop

When the flow passages in a packed column are assumed to be a bundle of tortuous channels the single-phase pressure drop can be expressed as

$$\Delta p = \lambda \cdot \frac{\tau \cdot H}{d_{eq}} \cdot \frac{\rho \cdot v^2}{2} \quad (1)$$

where Δp is the pressure drop in Pa, λ the friction factor of the tortuous channels, τ tortuosity factor to correct for greater path length, H the height of the packed bed, d_{eq} the equivalent hydraulic packing diameter, ρ the density of the fluid, and v the gas velocity in the channel. Since the superficial velocity, v_0 is equal to $v \cdot \epsilon$ and d_{eq} is equal to $4\epsilon/a$, where ϵ is the void fraction and a is the geometric area of the packing, the pressure drop can be expressed as

$$\frac{\Delta p}{H} = \lambda \cdot \tau \cdot \frac{a}{8\epsilon^3} \cdot \rho \cdot v_0^2 = \Lambda \cdot \frac{a}{6\epsilon^3} \cdot \rho \cdot v_0^2 \quad (2)$$

This equation is equivalent to Ergun's equation for flow through a bed of particles¹. The friction factor can be evaluated as

$$\Lambda = \frac{K_{lam}}{Re} + K_{turb} \quad (3)$$

Where Re is the Reynold's number defined as:

$$Re = \frac{6 \cdot v_0 \cdot \rho_G}{a \cdot \mu_G} \quad (4)$$

K_{lam} and K_{turb} for packings must generally be determined through experiments.

Several authors have presented Λ -values for single phase flow. In the books of Ramm² and Reichelt³ correlations are given for dumped and stacked rings, saddle packings, honeycomb blocks, and grids. In a recent work, Bemer and Kalis⁴ used the correlation of Ergun to estimate the dry pressure drop for Raschig rings. They introduced a correction factor, ϕ , defining an effective porosity and specific area. The following expression was evaluated for the turbulent region.

$$\frac{\Delta p}{H} = 0.29 \cdot \frac{a}{\phi^2 \cdot \epsilon^3} \cdot \rho \cdot v_0^2 \quad (5)$$

where $\phi = 0.6$ for Raschig rings and $\phi = 0.8$ for Pall rings.

2.2 Liquid Holdup

From reported liquid holdup data^{5,6} it seems evident that the gas flow does not affect the liquid holdup below loading. By using a channel model Bemer and Kalis⁴ could estimate the liquid holdup below loading in a bed of Raschig rings and Berl saddles from

$$h = C_1 \cdot a \cdot (\mu_L / \rho_L)^{2/3} \cdot Re_L^{2/3} \quad (6)$$

where $C_1 = (f/32 \cdot g)^{1/3}$, is a constant describing the alternating constrictions and enlargements of the flow channels. This constant was evaluated from experiments. The liquid Reynold's number is defined as $Re_L = \frac{4 \cdot L}{a \cdot \mu_L}$ where L is the liquid flow rate.

A model of inclined surfaces for predicting liquid holdup was proposed by Davidson⁷, and later modified by Buchanan⁸. At higher liquid Reynold's number, where the inertia forces predominates, Buchanan proposed that the liquid loses a fraction of its kinetic energy at the end of each packing piece. By using a combination of viscous film flow and this gravity-inertia model a good agreement with experimental data for Raschig rings was established. However, the model still involves empirical constants. Later, Hutton and coworkers⁹ introduced the effect of pressure gradient into Buchanan's model. In this way, liquid holdup above the loading condition could also be estimated.

2.3 Wet Pressure Drop

The wet pressure drop is generally expressed as the dry pressure drop multiplied by a factor due to the holdup. This factor could easily be obtained by substituting the free space of the wetted packing, $\epsilon' = \epsilon - h$, for the free space of the dry packing in equation (2). Thus for constant friction factor and negligible increase in relative velocity the wet pressure drop can be expressed as:

$$\frac{\Delta p_{\text{wet}}}{\Delta p_{\text{dry}}} = \left(1 - \frac{h}{\epsilon}\right)^{-3} \quad (7)$$

Bemer and Kalis⁴ considered the reduction of channel diameter and used this diameter in the pressure drop equation (equation 1) to obtain

$$\frac{\Delta p_{\text{wet}}}{\Delta p_{\text{dry}}} = \left(1 - \frac{h}{2 \cdot \epsilon}\right)^{-5} \quad (8)$$

This implies that the liquid film thickness is sufficiently small compared with the channel width. Often this is not so, which means that theoretically an exponent of -3 should be used instead of -5. For flow through slotted channels, as in the Plasdek packing, only equation (7) is theoretically justified.

However, both these channel models give a too low pressure drop. This is mainly due to liquid accumulation at contact points between the individual packing pieces. Bemmer and Kalis⁴ thus proposed a constriction model, where the pressure drop is determined by the change in kinetic energy at the contact points. They introduced a constriction factor, x , which relates the film thickness in the narrow part to the film thickness in the wide part of the channel as

$$\delta_N = x^{-2/3} \cdot \delta \quad (9)$$

where x must be determined experimentally.

Finally they could estimate the wet pressure drop from

$$\frac{\Delta p_{\text{wet}}}{\Delta p_{\text{dry}}} = \left(1 - \frac{h}{2 \cdot x^{5/3} \cdot \phi \cdot \epsilon}\right)^{-5} \quad (10)$$

With $x = 0.485$, for Pall rings, the wet pressure drop could be estimated to within 20 %.

Another way of estimating the pressure drop is through the generalized pressure drop correlation developed from Sherwood's flooding point correlation¹⁰ and reworked by Lobo¹¹, Leva¹², and Eckert¹³. In their latest design manual, the Norton Co. presents a revised form of the generalized pressure drop correlation, as displayed in figure 1. The packing factor, F , appearing in the ordinate group is considered to be a constant only dependent on the packing shape and size. It thus reflects the influence of friction and form resistance to the total flow resistance of the packing. However, some variation with the liquid flow rate has been observed. Kleinhückelkotten¹⁴, for instance, states that

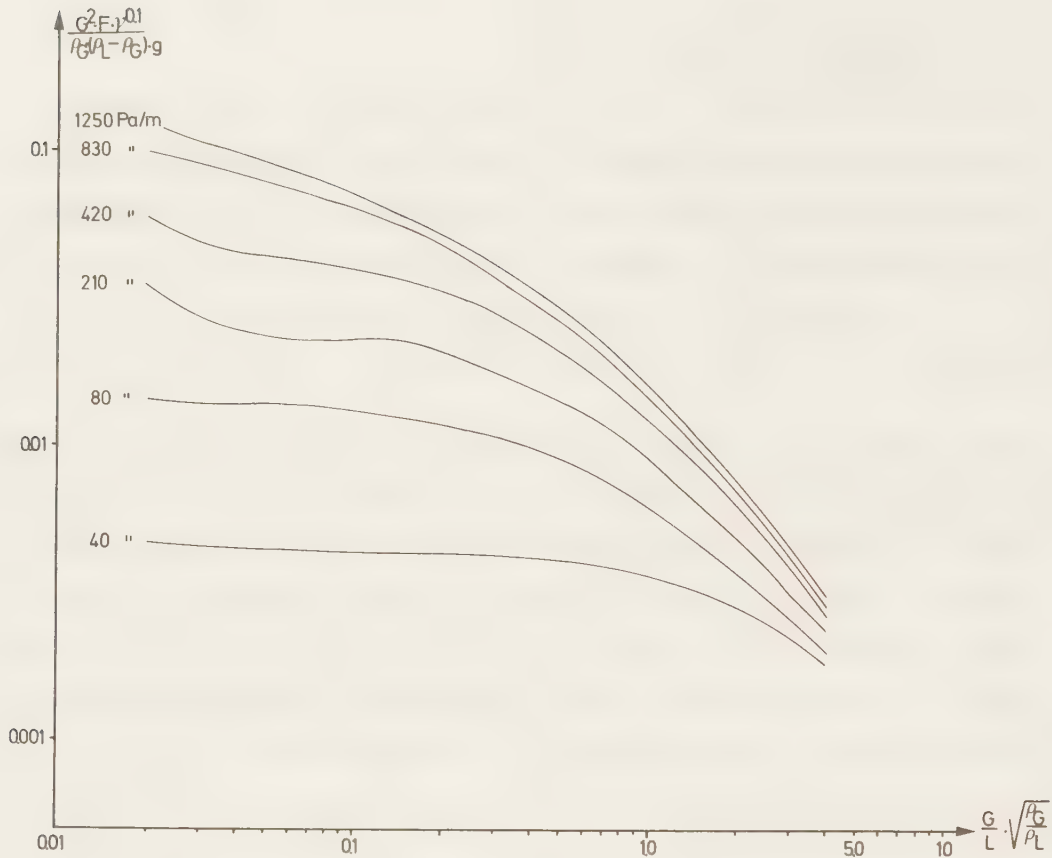


Figure 1. Modified generalized pressure drop correlation

the liquid flow rate dependency could be expressed as:

$$F_L = F_0 + A \cdot \ln \left(\frac{L}{G} \sqrt{\frac{\rho_G}{\rho_L}} \right) \quad (11)$$

where F_0 and A are evaluated from experiments.

A variety of other pressure drop correlations have been developed. Ramm summarizes several of them in his book "Absorption of gases"². Other examples are the works of Beck¹⁴, Reichelt¹⁵, and Teutch¹⁶.

2.4 Flooding

It is important to successfully predict the flooding condition in a packed column. The flooding point puts a limit to gas and liquid load handling capacity. Often the flooding point is the basis for dimensioning the cross sectional area of the tower. However, the flooding point is not a well-defined concept. Many investigators used the visual flooding point, others the graphical as obtained when plotting pressure

drop against gas velocity in logarithmic coordinates. Hutton et al⁹ state, in their discussion of flooding in packed beds, that the flooding point is reached when the derivative of liquid flow rate with respect to liquid holdup becomes zero at a constant gas rate. Often it is only parts of the packing that are flooding, making the phenomenon even less definable.

The most widely used flooding point correlation is that developed by Sherwood, Shipley and Holloway¹⁰. As earlier mentioned, this correlation is also the basis for the generalized pressure drop chart. The flooding point correlation as revised by Leva¹² and presented by Perry⁵ is shown in figure 9. The dimensionless groups on the axes are obtained from dimensional analysis, except for the viscosity term. The abscissa value thus describes the square root of the kinetic energy ratio of the two phases. Likewise the ordinate group is the ratio of friction force to force of gravity. For the packing characteristic, a/ϵ^3 , Lobo¹¹ have introduced the packing factor, also appearing in the generalized pressure drop correlation. By introducing a packing factor, account can be taken of different form and friction resistance for different packings. Another way of doing this was proposed by Zhavoronkov¹⁸, as cited by Ramm². Zhavoronkov used an ordinate value equal to the ratio of resistance of dry packing, Δp_{Dry} , to the weight of liquid per square meter of cross section;

$$\text{Ordinate} = \frac{\Delta p_{Dry}}{\rho_L \cdot g \cdot H} = \Lambda \cdot \frac{a}{6\epsilon^3} \cdot \frac{\rho_G \cdot v_o^2}{\rho_L \cdot g} \quad (12)$$

The friction factor, as determined from dry pressure drop experiments, will allow for different flow resistance of different packings. However, it is not likely that the same variables that govern the pressure drop also govern the flooding phenomenon; at flooding gas is merely bubbling through a liquid layer rather than flowing around the packings.

Mersmann¹⁹ developed a flooding point correlation from the model of a thin liquid film flowing in a channel. By assuming the flow to be laminar he obtained two dimensionless groups $\Delta p/H \cdot \rho_L$ and δ/d_h , from which a correlation under turbulent conditions could be derived. Since δ/d_h is a function of liquid irrigation rate and $\Delta p = \Delta p_{Dry} + \Delta p_{Wet}$ the final correlation was expressed as:

$$\frac{\Delta p_{Dry}}{H \cdot \rho_L} = f \left\{ \left(\frac{v_L}{g} \right)^{1/3} \cdot \frac{L}{\rho_L} \cdot \frac{(1-\epsilon)}{d_r \cdot \epsilon} \right\} \quad (13)$$

$$\text{where } d_r = \frac{6 \cdot \text{Volume of the packing}}{\text{Area of the packing}} = \frac{6 \cdot (1-\epsilon)}{a} = \frac{6 \cdot (1-\epsilon)}{4 \cdot \epsilon} \cdot d_{eq} \quad (14)$$

Later on Reichelt^{3,16} modified this expression by instead using a hydraulic diameter defined as

$$d_{h,w} = d_{eq} \cdot \frac{1}{1 + \frac{2}{3 \cdot (1-\epsilon)} \cdot \frac{dr}{D}} \quad (15)$$

where D is the diameter of the tower. In this way pressure drop measurements from small columns, where wall effects may be significant, could also be incorporated in the correlation. Reichelt's definition of the hydraulic diameter also means that $d_{h,w} = D$ for an empty tower.

The flooding point correlation of Mersmann¹⁹ and Reichelt^{3,16} is somewhat different from the Sherwood correlation. By the former method it is possible to predict the flooding velocity for every liquid load from dry pressure drop measurements. The Sherwood correlation needs the ratio between gas and liquid flow rates to predict the flooding velocity. This is not a major drawback, however, because when designing a packed column the total gas flow is normally known. The corresponding liquid flow rate is then for instance evaluated from equilibrium considerations. In the Mersmann-Reichelt correlation the dry pressure drop must be estimated, for instance as earlier described. This introduces an additional uncertainty to the flooding point prediction.

There are still other flooding point correlations. Reichelt¹⁶ suggests that the Reynold's number for the gas phase should be plotted against the liquid Reynold's number. By suitable correction factors a good correlation was established for Raschig rings. Another correlation proposed by Eduljee²⁰ was recommended by Ramm². Eduljee plots

$$P \cdot \frac{v_o^2}{g \cdot d_{nom}} \cdot \left(\frac{1}{\frac{v_o \cdot d_{nom} \cdot \rho_G}{\mu_L}} \right)^{0.1} \cdot \left(\frac{\rho_w}{\rho_L} \right)^2 \cdot \left(\frac{\rho_G}{\rho_{Air}} \right)^{0.82} = f \left(\frac{L}{G} \cdot \frac{\rho_G}{\rho_L} \right) \quad (16)$$

where P is an experimentally determined packing constant and d_{nom} the nominal packing diameter. Flooding points for Raschig rings, Pall rings

Intalox saddles, and Tellerettes were predicted in this way. Eduljee also included the loading line in his graph. It is thus possible to directly establish the operating condition.

3. EXPERIMENTAL

The pressure drop for the three packings were measured in a pilot-plant described elsewhere²¹. The inside tower diameter was 0.3 meter and the packing height could be varied up to 3.5 meters. The ratio of tower diameter and nominal packing size was greater than 10/1, which ensured that wall effects could be neglected. Pressure taps were located immediately above and below the packed section. When a liquid and gas flow rate had been adjusted and stabilized over ten to fifteen minutes, the pressure drop was registered by a pressure transducer. Care was taken that the gas flow was increased during each test series, avoiding hysteresis effects. The measured pressure drops were corrected for pressure drop in the gas injector.

Pressure drops were measured at three different heights: 0.75, 1.5, and 3 m. The Tellerettes and the Intalox saddles were packed dry in a random fashion. They were packed densely, with more packing pieces per cubic meter tower volume than recommended by the manufacturers, as is evident from table 1.

Packing	nominal size m	$\frac{a}{m^2/m^3}$	ϵ %	Pieces/m ³
Intalox saddles	0.025	207	91	72000
Tellerettes	0.025	180	87	41000
Plasdek	-	225	97	5 blocks of 0.3 m under right angles

Table 1. Packing characteristics

This was accomplished by pressing during the packing procedure. A test series with the recommended number of pieces of Intalox saddles was also performed. The Plasdek packing was manufactured in whole blocks equal to the tower diameter and with a height of 0.3 m. The blocks were placed upon each other with the sheets at right angles. The blocks had no drainage tips.

Liquid holdup measurements were also performed. This was accomplished by measuring the amount of liquid which has disappeared from the liquid

tank below the tower. The liquid volume in the pipes and distributor was subtracted from this volume. All measurements were made at zero gas flow. Sufficient drainage time was permitted between each liquid flow to establish the dynamic liquid holdup. It should be pointed out that this method of establishing the liquid holdup is rather inaccurate; only a rough measure of the holdup is established.

4. RESULTS

The resulting pressure drops for the three packings are presented in figures 2, 3, and 4. In these figures the pressure drop is plotted as a function of gas velocity in logarithmic coordinates with the liquid load as parameter. The pressure drops presented were all measured at a packing height of 1.5 meter. About the same pressure drops were obtained at all three heights (± 20 Pa/m). The small variations were attributed to end effects at a packing height of 0.75 m and to maldistribution of the liquid at a packing height of 3 m.

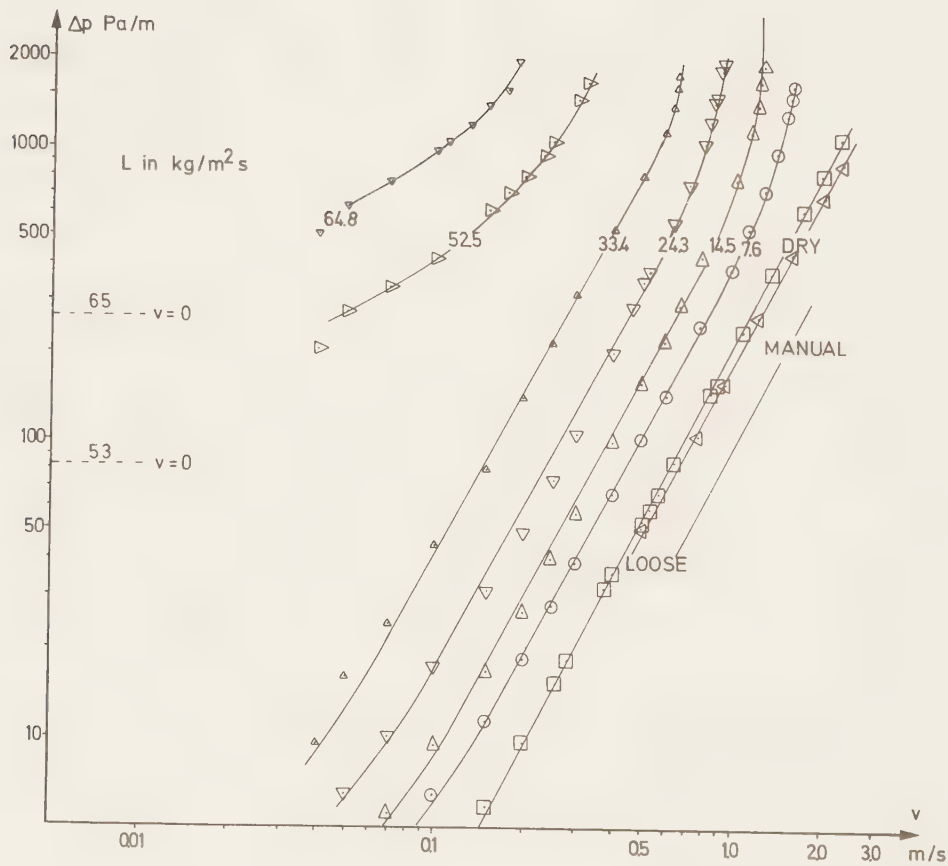


Figure 2. Pressure drop - Intalox saddles

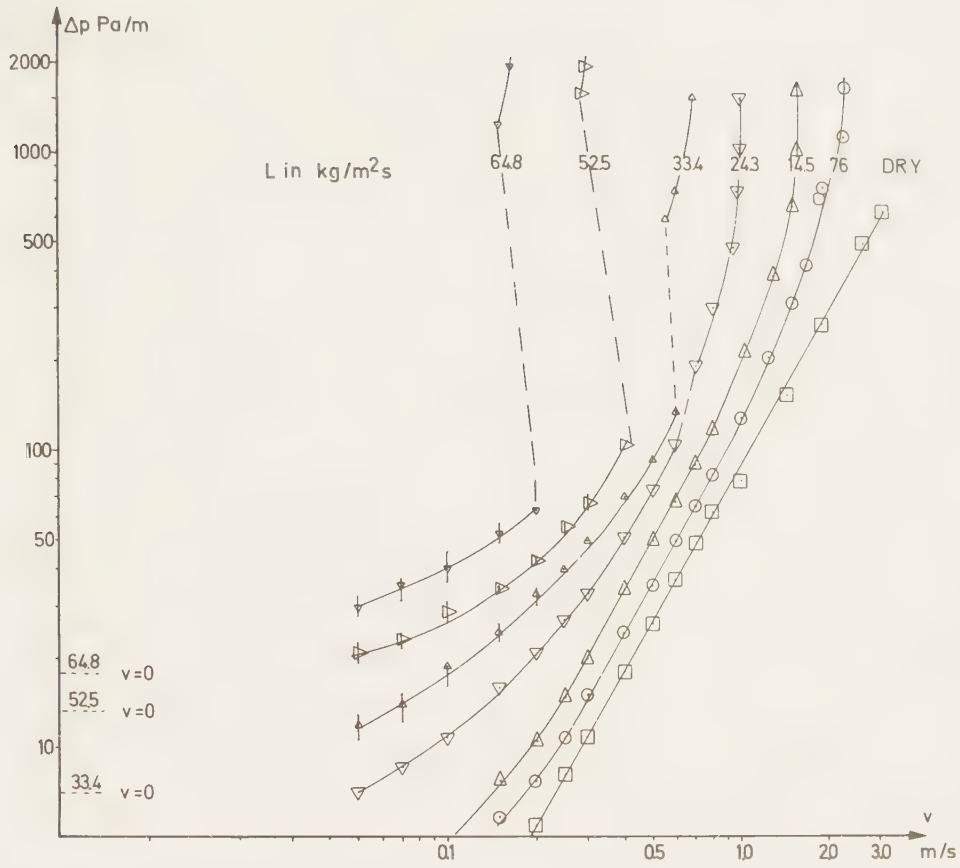


Figure 3. Pressure drop - Tellerettes

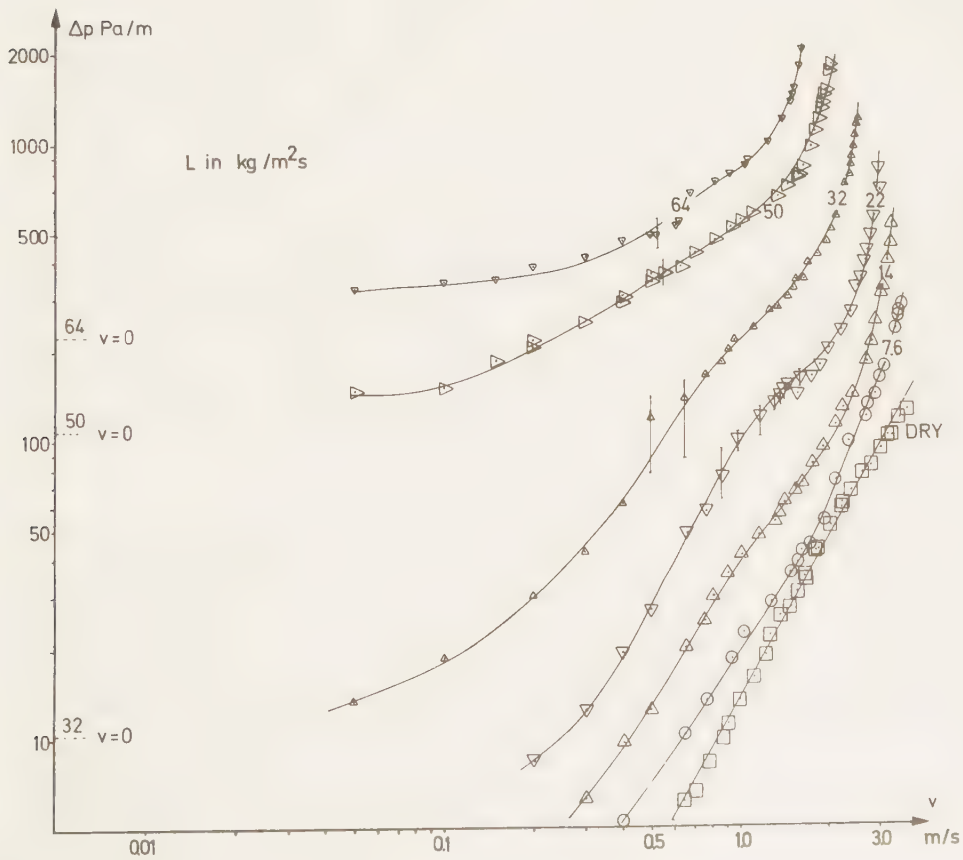


Figure 4. Pressure drop - Plasdek

The three packings exhibit dry pressure drop lines with different slope, as presented in table 2.

	Intalox Saddles	Tellerettes	Plasdek
Slope	1.92	1.75	1.70

Table 2. Slope of dry pressure drop line

At moderate liquid loads essentially straight lines are obtained. For the Intalox saddles and the Tellerettes they are parallel to the dry lines, indicating the influence of liquid holdup. The line for the Plasdek packing has a slope less than the slope for the dry packing. This indicates the more pronounced effect of the change in relative velocity between the phases. The plateaus at somewhat elevated gas loads seem to indicate an increase in roughness at the interface between the phases, for instance by wave formations. At sufficiently high gas load the pressure drop curves are vertical and the flooding point is reached. No sharp transition is observed at flooding. The loading point is not detectable because of a gradual change in pressure drop with gas load. At high liquid loads the pressure drop curves have a slope that is less than the dry line slope for each packing. The high relative velocity results in high interfacial stresses which induce axial mixing of the gas. Even at zero gas flow a pressure gradient is built up. Both pressure drop at zero gas flow and the induced concurrent gas flow at higher liquid flow rates have been measured for the Plasdek packing²². As seen from figure 2, 3, and 4 there are strong indications that the axial back-mixing in a tower filled with Plasdek is greater than in a tower filled with Intalox saddles or Tellerettes.

The pressure drop at flooding is somewhat different for the three packings. The Intalox saddles have a flooding pressure drop between 1500-2000 Pa/m, independent of liquid flow rate. The Tellerettes seem to flood at somewhat lower pressure drop, approx. 1000 Pa/m. With liquid flow rates above $20 \text{ kg/m}^2 \cdot \text{s}$ a tower with Tellerettes operates very unstably. Even at a relatively low gas flow rate the tower seems to reach the flooding condition. The pressure drop increases considerably and the gas flow starts to oscillate. A new, much higher,

pressure drop is established at a somewhat lower gas velocity. All packings show hysteresis effects between loading and flooding, but the Tellerettes seems to be more sensitive to whether the gas flow is increased or decreased. The Plasdek has a very low flooding pressure drop at low liquid flow rates, only about 300-500 Pa/m. From the sales manual it is obvious that the flooding point varies with the arrangement of the contact zone between the packing blocks. For instance, when the contact zones are arranged with drainage tips, higher flooding velocities are obtained. This indicates that the flooding starts primarily at the contact zones.

A comparison of the pressure drop obtained for the three packings reveals that the Plasdek has a very low pressure drop compared with Intalox saddles and Tellerettes. The Tellerettes, on the other hand, have a somewhat lower pressure drop than the Intalox saddles. The liquid load influences the pressure drop of the three packings differently. The Tellerettes have a very low flooding point at liquid loads above $20 \text{ kg/m}^2 \cdot \text{s}$, where the Tellerettes can only be used at low gas flow rates. The Plasdek is also more dependent on liquid flow; at increasing liquid flow rates the increase in pressure drop is relatively greater than for the same increase in a bed of Intalox saddles. Still the pressure drop is lower in a bed of Plasdek and also higher gas loads are permitted before flooding.

The packing factor, F , introduced by Leva in the generalized pressure drop correlation can be used as a measure of the capacity of different packings. A low packing factor means a high capacity at a given pressure drop or a low pressure drop at a given capacity. The packing factors can be estimated by matching measured pressure drops to the relevant pressure drop lines as presented in figure 1. Table 3 presents such packing factors at some different pressure drop levels. As is seen from the table the Plasdek permits about 2-4 times as high a gas load as Intalox saddles at the same pressure drop. Likewise, the Tellerettes have a forty percent higher capacity than the Intalox saddles. It is also seen that when the Intalox saddles are packed loosely, i.e. with the recommended number of packing pieces per cubic meter, a lower packing factor is obtained. However this packing factor is still about twice as large as reported by the manufacturer.

ΔP Packings	F_m^{-1}					
	1250	830	420	210	80	40
Intalox Saddles Dense	260-230	280-260	300-310	320-370	480-460	450-550
Intalox Saddles Loose	210-180	210-180	220-200	250-200	340-300	-
Tellerettes	150 Flooding	150-160	150-170	140-170	180-170	160-240
Plasdek	55 Flooding	50-60 Flooding	30-40	20-40	30-100	20-100

Table 3. Packing factors

The resulting liquid holdups for the three packings are plotted in figure 5.

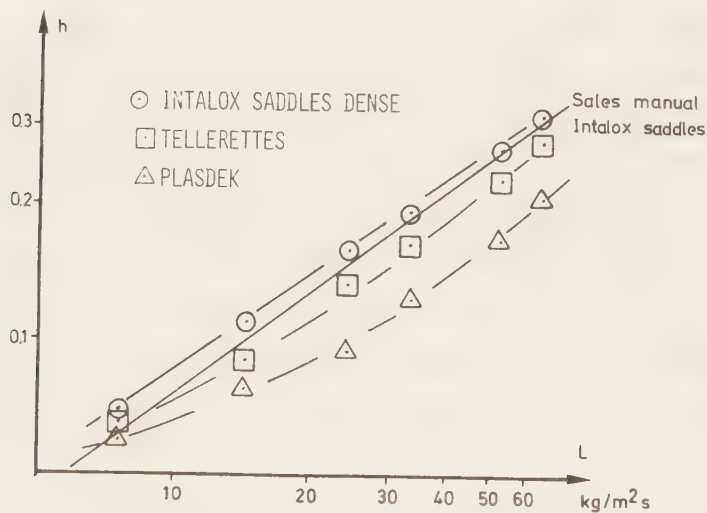


Figure 5. Liquid holdup

The general trend is a lower liquid holdup in the Plasdek packing than in a bed of Tellerettes or Intalox saddles. Also a somewhat higher liquid holdup was obtained with the dense packed Intalox saddles compared with the sales manual. The decreasing slope at lower liquid flow rates for the Tellerettes and the Plasdek seems to indicate a higher static holdup in a bed of these packings than in a bed of Intalox saddles. It should, however, once more be pointed out that the method used to determine the holdup could be rather inaccurate.

4.1 Prediction of Pressure Drop in Packed Columns

As earlier mentioned, there are basically two ways of predicting the pressure drop in a packed column. One method is to use the generalized pressure drop correlation. When using the other method the dry pressure drop must first be predicted. Together with the liquid holdup, the wet pressure drop can then be estimated. The first method predicts pressure drops from below loading to flooding, while the second one can only be used to predict pressure drops below and up to loading.

The generalized pressure drop correlation is presented in figure 1. When used together with the packing factors in table 3 pressure drops can be estimated for different gas and liquid loads at different pressure drop levels. However, the problem of estimating the pressure drop has now become a problem of estimating the packing factor as a function of liquid load and pressure drop level. Table 3 clearly shows that F varies considerably. The two limits given at each pressure drop level is the F -value obtained at the lowest and highest liquid flow rate, generally also expressing the total variation by L . Using a mean value of F will cause considerable deviations of predicted pressure drops from measured ones, depending on the value of the abscissa. The deviation may very well be around a hundred percent, especially at high abscissa values.

In its present form, the generalized pressure drop correlation seems to be of little value in predicting pressure drops for any of the three packings examined. Even for Intalox saddles, one of the packings for which the correlation was proposed, the precision is insufficient. It is also not likely that the correlation should hold for the Plasdek, since this packing has a very low flooding pressure drop at a low liquid flow rate. It is thus impossible to obtain a pressure drop of 1250 Pa/m without passing the flooding point. An F -value that mainly describes the flow situation below flooding cannot be expected to describe the bubbling phenomena above flooding.

The pressure drop lines in figure 1 may perhaps be further changed to give better pressure drop estimations at a constant F -value. However a much greater number of pressure drop measurements must be used to establish the empirical type of relationship.

From the dry pressure drop curves in figure 2-4 the empirical constants appearing in the dry pressure drop models can be evaluated. The ϕ -values proposed by Bemer and Kalis⁴ are determined from the slope of the dry pressure drop curves with the least square method. With a ϕ -value of 0.68 for the Intalox saddles, 1.15 for the Tellerettes and 3.04 for the Plasdek the dry pressure drop is correlated as presented in figure 6. The model is good at high pressure drops but tends to give low values at lower pressure drops.

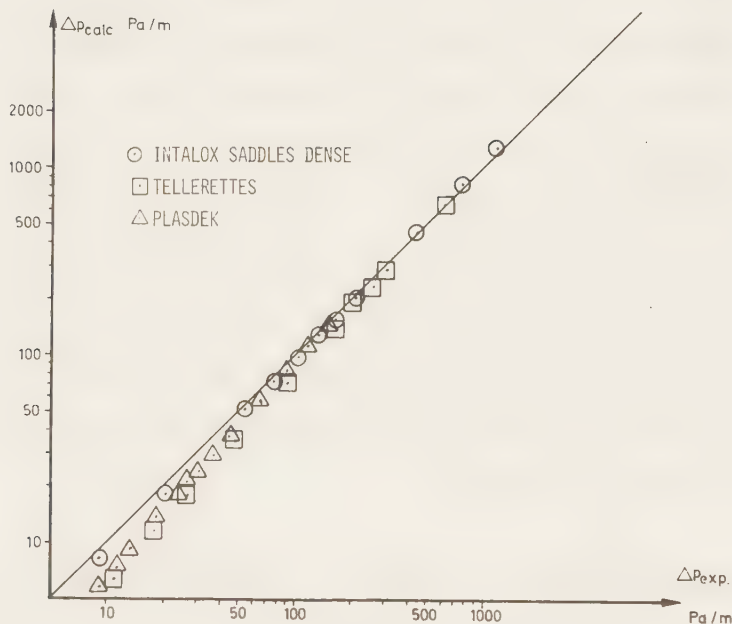


Figure 6. Dry pressure drop correlation - Bemer and Kalis⁴

It is interesting to note the magnitude of the ϕ -values for the three packings. Since they are greater than 1 for the Tellerettes and the Plasdek, they simply express the diminished form resistance imposed by different packings compared with the spheres used in Ergun's relationship¹. The sphericity factor included in Ergun's equation¹ does not entirely take account of different flow separation and wake formation in a bed of packings.

The relatively poor correlation obtained by the method of Bemer and Kalis⁴ at low pressure drops, can be attributed to that the flow is not fully turbulent. The laminar term in Ergun's equation¹ could thus

not be omitted. However, the prediction is rather poor even if this term is included. As is seen from figure 7 the Λ -values for the packings do not attain a constant value even at the highest Reynold's number. This particular Reynold's number means a velocity in the upper range of operation of a packed column.

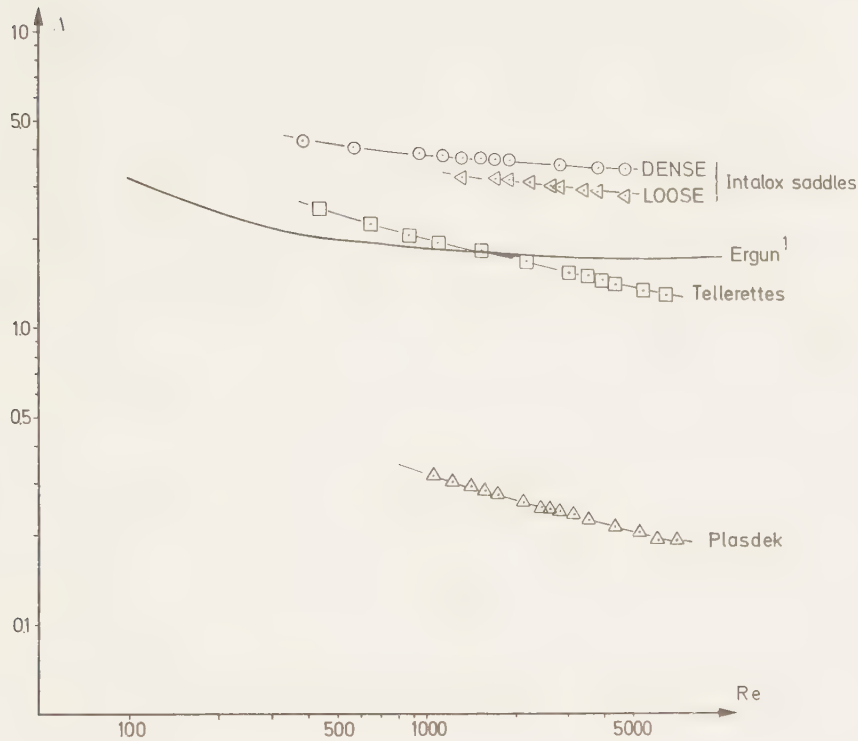


Figure 7. Dry pressure drop correlation - Ergun¹

The Ergun type of correlation¹ can not adequately describe the single phase flow in a bed of commercial packings. Generally, a better correlation would be obtained if equation (3) is rewritten as

$$\Lambda = \frac{K_{lam}}{Re} + \frac{K_{turb}}{Re^\alpha} \quad (3')$$

By using the least square method the values of K_{lam} , K_{turb} , and α can be obtained as presented in table 4. By using these constants in equation (3') and (2) the dry pressure drop can be correlated for the three packings with a maximum error of 2 percent.

Packing	α	K_{lam}	K_{turb}	Standard Deviation in Δ -prediction
Intalox Saddles	0.07	47.2	6.4	0.017
Tellerettes	0.20	130.8	7.5	0.014
Plasdek	0.25	21.4	1.7	0.0014

Table 4. Correlation of Δ -values

The measured liquid holdup could be correlated as proposed by Bemmer and Kalis⁴. The resulting relationship is valid for flow conditions up to loading. They may also, according to Bemmer and Kalis⁴, be used for predicting liquid holdups with other liquids and with other sizes of the packings. The empirical constant, C_1 , in equation (6) is evaluated by the leastsquare method. The resulting constants are presented in table 5. With these values the experimental holdups are correlated within 10 per-cent as shown in figure 8.

Packing	C_1	s	f	range of Re_L
Intalox saddles	0.13	0.006	0.69	150-1250
Tellerettes	0.12	0.007	0.54	170-1400
Plasdek	0.08	0.007	0.16	130-1150

Table 5. Correlation of liquid holdup

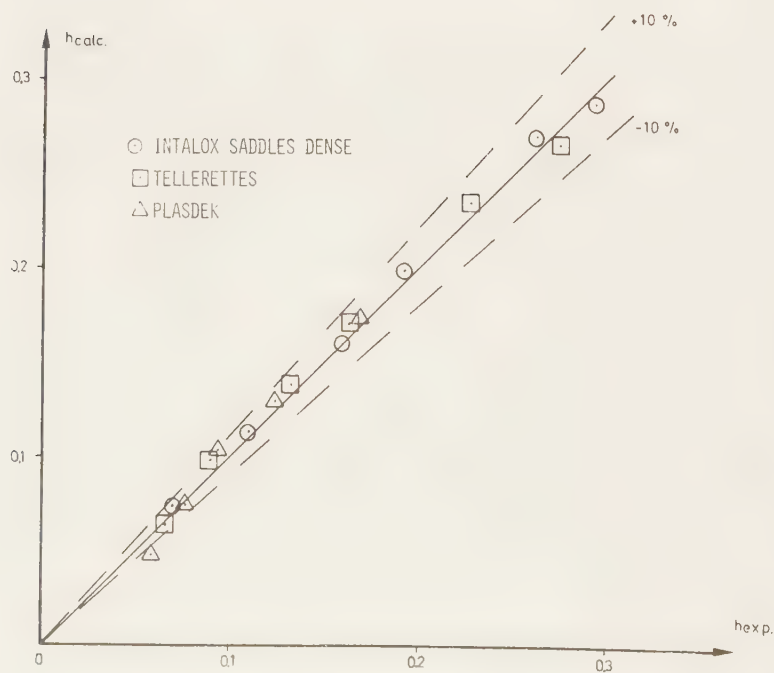


Figure 8. Correlation of liquid holdup

In table 5 friction factors for the liquid flow in the packings are also presented. As is seen from these friction factors, the flow of liquid in the packings are of turbulent character, except for the Plasdek at $8 \text{ kg/m}^2 \cdot \text{s}$. For this point the Nusselt film relation could equally well have been used.

The wet pressure drop can now be correlated with the use of equation (10). The constriction factor, x , appearing in the equation has been evaluated with the least square method for the three packings. In table 6a the established constriction factors are presented together with the standard, mean, and maximum deviations, and the flow regime within which the model was tested.

Packing	Constriction factor, x	Deviation			Flow regime	
		Max %	Mean %	s Pa	L $\text{kg/m}^2 \cdot \text{s}$	G $\text{kg/m}^2 \cdot \text{s}$
Intalox saddles	0.62	15	9	30	7.6	0.2-1
					14.5	0.2-1
					24.3	0.2-0.7
Tellerettes	0.55	25	10	12	7.6	0.2-1.5
					14.5	0.2-1.2
					24.3	0.3-0.6
					33.4	0.3-0.5
					52.5	0.3-0.5
Plasdek	0.24	50	18	8	7.6	0.6-3
					14.0	0.6-2.5

Table 6a. Constriction factors for wet pressure drop

$$\zeta = \left(1 - \frac{h}{x \cdot \phi^2 \cdot \varepsilon}\right)^{-5}$$

The experimental pressure drops have also been correlated with equation (7) including an additional constriction factor. As is seen from table 6b, about the same accuracy was obtained as when equation (10) was used.

It can thus be concluded that by using constriction factors for the packings together with the models for dry pressure drop and liquid hold-up, the wet pressure drop can be correlated to within 20 %. Only the pressure drop for the Plasdek are correlated with less accuracy, especially as the liquid load increases. From the pressure drop curves (fig. 4) it might be suspected that the liquid holdup is not the only parameter that influences the wet pressure drop. Furthermore, a higher influence of liquid load could be noticed in spite of the smaller in-

crease in holdup compared with the Tellerettes and Intalox saddles.

Packing	Constriction factor, β	Deviation			Flow regime	
		Max %	Mean %	s Pa	L kg/m ² s	G kg/m ² s
Intalox saddles	0.59	13	8	21	7.6	0.2-1
					14.5	0.2-1
					24.3	0.2-0.7
Tellerettes	0.47	14	9	15	7.6	0.2-1.2
					14.5	0.2-1
					24.3	0.3-0.6
					33.4	0.3-0.5
Plasdek	0.083	48	18	8	7.6	0.6-3.0 0.6-2.5

Table 6b. Constriction factors for wet pressure drop

$$\zeta = \left(1 - \frac{h}{\beta \cdot \phi^2 \cdot \epsilon}\right)^{-3}$$

The ultimate purpose of the proposed correlation is to be able to predict pressure drops at relevant working conditions. This means that pressure drops with other liquids and gases as well as for the packings at other sizes are to be predicted. This has not been tested, because of lack of data. The result of Bemer and Kalis⁴, however, indicates that wet pressure drop can be estimated with acceptable accuracy with the procedure outlined here. It should also be pointed out that the wet pressure drop at high liquid loads, where the gas-phase tends to be back-mixed, can not be predicted with this simple model. Likewise, the model is only valid below loading, because of the strong influence of gas velocity on liquid holdup above loading. With the present state of knowledge it seems preferable to use experimentally determined pressure drop data.

4.2 Prediction of Flooding in Packed Columns

The experimentally found flooding points have been compared with flooding points predicted with some of the existing flooding point correlations. The graphical flooding point was used here. When the experimental flooding points are plotted as originally proposed by Sherwood et al¹⁰ figure 9 is obtained. It is apparent that the flooding points for the Intalox saddles and the Tellerettes can be predicted satisfactorily from the curve for dumped packings. The Plasdek, on the other hand, will permit

a higher gas load before flooding. The flooding points for the Plasdek fall somewhere between the curves for dumped and stacked packings.

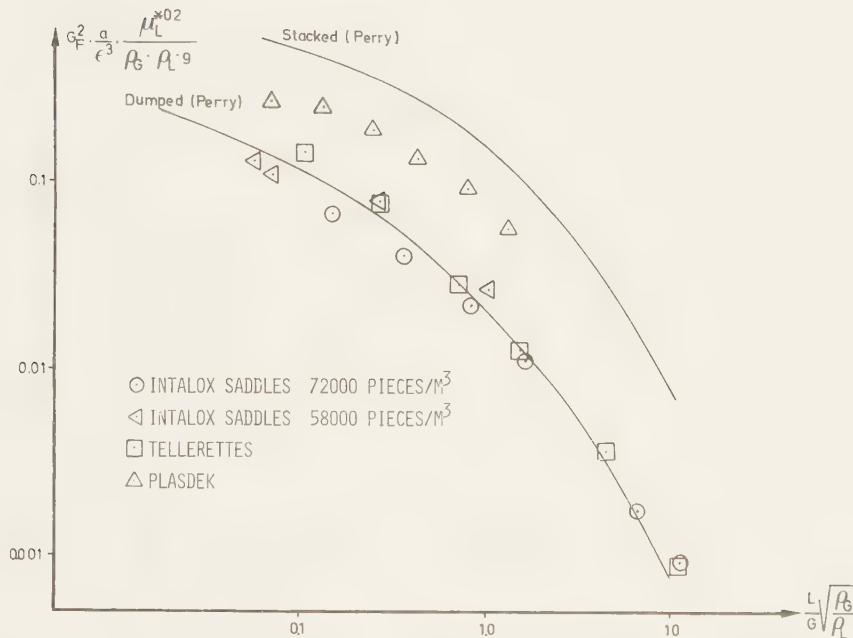


Figure 9. Flooding point correlation - Perry⁵

To obtain the flooding points in the figure, a/ϵ^3 was used as proposed by Sherwood¹⁰. This may also be the most logical parameter to use to take account of the influence of packing geometry. A low void means, for instance, narrow sections which results in higher velocities and interfacial stresses. These narrow sections at the contact points between the packings are more easily filled with liquid, which means flooding. For the Plasdek, the most critical section, where the flooding first starts, will be at the contact zones between successive blocks. At low liquid loads it seems likely that the bed is flooding only at these zones. This means a very low flooding pressure drop. Another consequence is that in this region the flooding velocity is almost independent of liquid load. The flooding curve will thus be almost horizontal at low abscissa values. For these reasons it seems unlikely that one flooding point correlation can be used for all packings without taking account of the specific flooding characteristics of each packing. Results of the same sort were found by Choi et. al.²⁴

When the experimental flooding points are plotted according to Zhavoronkov¹⁸, as in figure 10, one curve is obtained for each packing.

This reflects the magnitude of the dry pressure drop for the three packings.

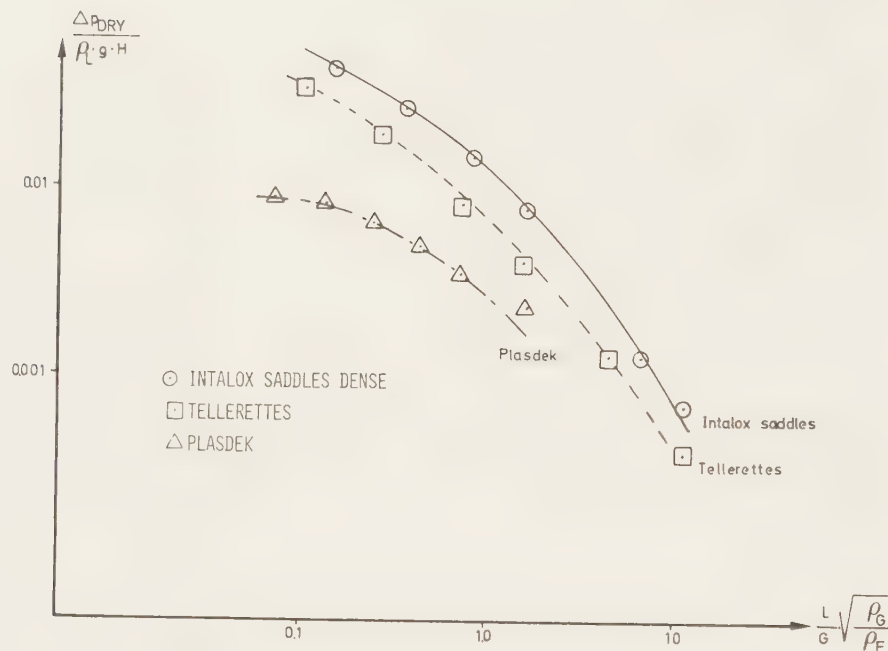


Figure 10. Flooding point correlation - Zhavoronkov¹⁸

The correlation proposed by Mersmann¹⁹ and Reichelt¹⁶ also uses the dry pressure drops as ordinate values. The abscissa only contains liquid quantities. By this method, three flooding lines are also obtained, as is obvious from figure 11. The flooding points for the Intalox saddles coincide well with the flooding line as established by Reichelt for a variety of packings including the Intalox saddles.

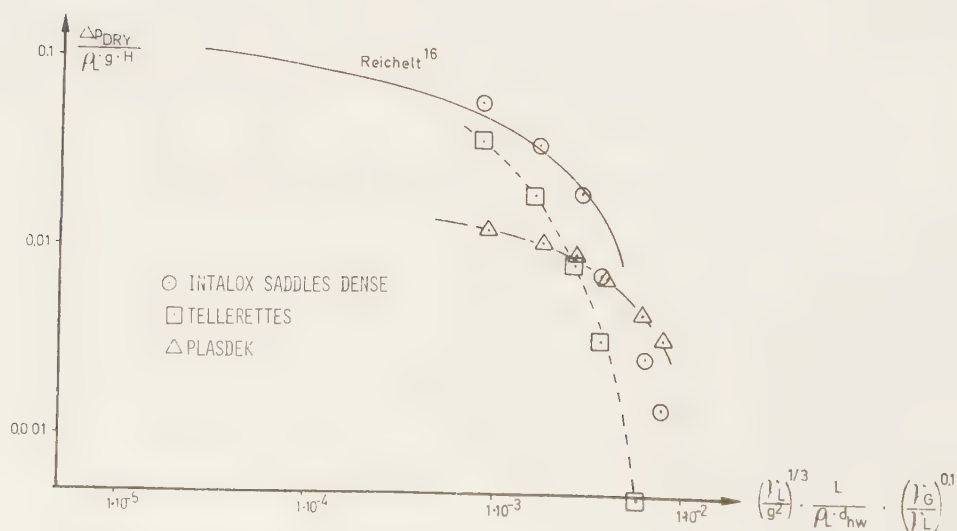


Figure 11. Flooding point correlation - Mersmann¹⁹, Reichelt¹⁶

Finally, the flooding points are correlated by the method proposed by Eduljee in figure 12.

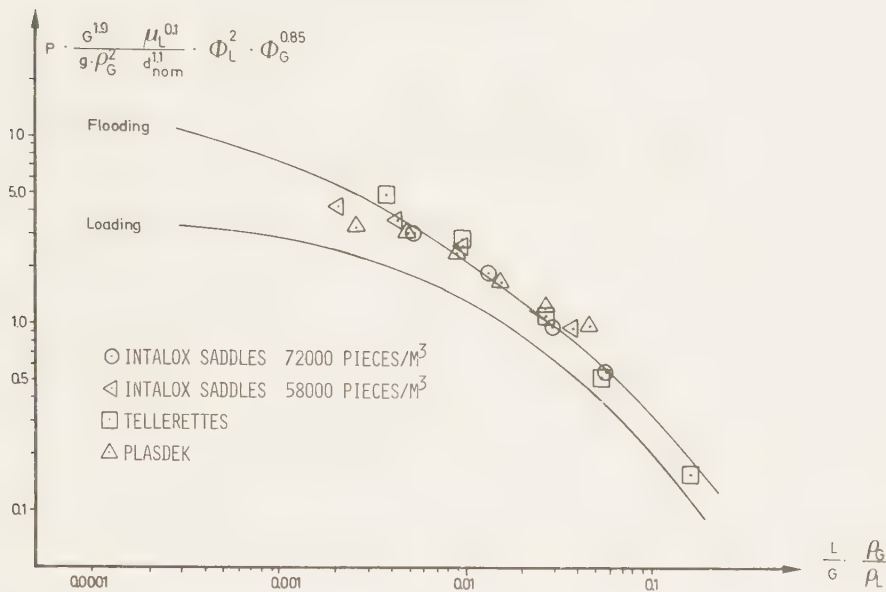


Figure 12. Flooding point correlation - Eduljee^{2,1}

The packing factors, P , were determined as the mean values of the factors that each ordinate value has to be multiplied with in order to fall on the graph of Eduljee. Since the graph presented by Eduljee is not dimensionless, the relationship as reworked by Ramm² (eq. 16) was used. Furthermore, the packing factors obtained by Eduljee were not dimensionless, a fact overlooked by Ramm. After suitable conversions dimensionless packing factors as displayed in table 7 are obtained.

Packing	P	$P_{Eduljee}$
Intalox saddles loose	0.21	0.21
Intalox saddles dense	0.28	
Tellerettes	0.23	0.23
Plasdek	0.075	

Table 7. Packing factors - method of Eduljee

A close resemblance is obtained with the packing factors presented by Eduljee. By using the packing factors in table 7 there was some scatter of flooding points for the three packings as displayed in figure 12. The best correlation was found for the Intalox saddles. It seems, however, somewhat unrealistic that the nominal diameter of the packing

should be able to describe the influence of type and dimension of the packing. For the three types of packing examined here the nominal diameter is rather a measure of the geometrical area and has, more or less, no physical meaning.

The flooding points for the three packings can be made to coincide with one flooding point curve by the introduction of a "flooding" factor. This "flooding" factor can be compared with the packing factor, but merely describes the distribution of contact points in the bed. The basis for the flooding factor is the flooding point curve for dumped packings (see figure 9). The ratio between the experimental ordinate value, using a/ϵ^3 , and the value obtained from this curve, at the same abscissa value gives the flooding factor after multiplication with a/ϵ^3 . Table 8 summarizes the resulting flooding factors.

Intalox saddles		Tellerettes	Plasdek
Loose	Dense		
270	310	245	90

Table 8. Flooding factors

These flooding factors vary with L/G . The values shown are mean values. By slightly changing the flooding point curve, according to ordinate values calculated with the mean flooding factors, this L/G dependency can be avoided. In figure 13 this "new" flooding point curve is plotted.

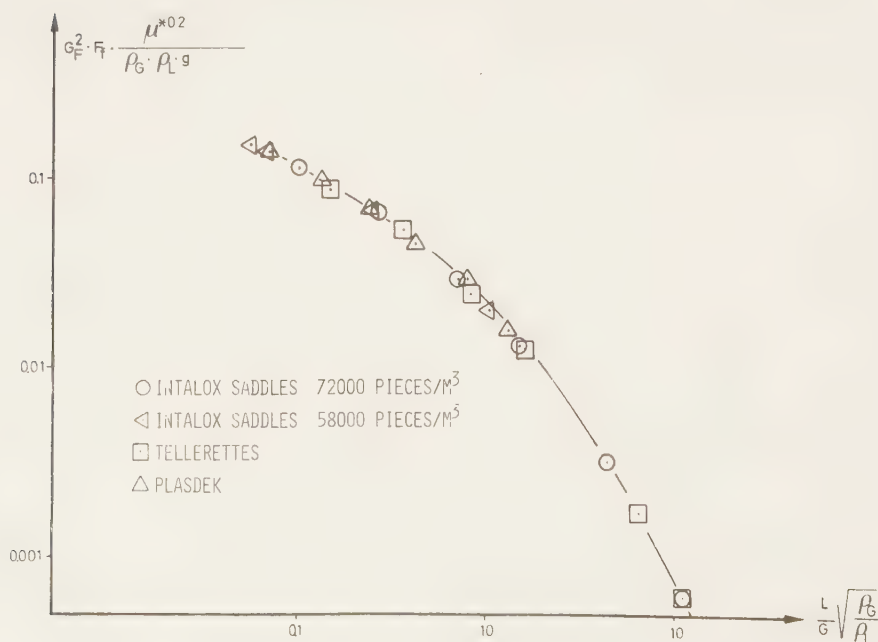


Figure 13. Flooding point correlation with flooding factors

However, the number of points is rather limited and a much more comprehensive work must be done to establish both the flooding factors and the flooding point graph. Similarly, flooding properties of other gases and liquids must be investigated. The influence of tower diameter and tower height should also be examined. Since only three packings, although quite different, were investigated the flooding point graph as presented by Perry⁵ can still be considered to be the most general one, but with the restriction that it is only valid for dumped packings. Finally, it may be noted that although the prediction of the flooding gas load seems rather inaccurate, the use of an arbitrary reduction factor (e.g. 0.5-0.6) to get the loading velocity means that the result still gives a rather safe operating condition. Strong indication of this is found from the pressure drop curves presented.

5. CONCLUSIONS

Pressure drop data are essential when designing packed towers. From the pressure drop data the diameter of the tower is determined. The three packings examined are believed to have a very high capacity. The result shows, a considerable variation in capacity. From the result, the following general conclusions can be drawn.

1. The Plasdek has the highest capacity of the three packings. The pressure drop is considerably lower at the same flow rates, and higher gas loads are permitted at every liquid load before flooding. This means a smaller tower diameter.
2. A bed of Tellerettes operates very unstably at higher liquid loads. When the liquid load is higher than $20 \text{ kg/m}^2 \cdot \text{s}$ the packing can only be used when the gas load is very low.
3. Higher pressure drops were obtained for the Intalox saddles than given by the manufacturer. This was partly because the bed contained more packing pieces per cubic meter than recommended. With the recommended number of pieces the pressure drop was still about 40 % higher.
4. At sufficiently high liquid loads, the gas phase is back-mixed, especially in a tower with the Plasdek packing. Generally this means a diminished driving force for the mass transfer.
5. The wet pressure drop is best correlated with a model based upon the interaction of liquid holdup on dry pressure drop in convoluted channels. The generalized pressure drop correlation can not be used for the estimation of pressure drop. It remains to be seen whether these correlations can be used for the estimation of pressure drop with other gases and liquids, and different dimensions of the packings.
6. The flooding point correlation proposed by Sherwood et. al.¹⁰ was considered the most general one. However, with a/ϵ^3 as the packing parameter, this correlation can only be used for dumped packings; the flooding points for the Plasdek fall between the lines for dumped packings and stacked packings. By the introduction of a "flood-

ing" factor a better correlation may be obtained. This must still be further tested.

7. The pressure drop correlations available today seem to be rather inaccurate. The correlations may be used to estimate the tower diameter, but for the exact pressure drop determination experimental data should be used.

Acknowledgements

The author wish to express his gratitude to Mr. N.-E. Carlstedt for excellent experimental work. For providing the examined packings Thurne Teknik, Ceilcote AB, and AB Carl Munters are gratefully acknowledged.

NOMENCLATURE

A	Constant for flow dependency of the packing factor	-
a	Specific geometric packing area	m ² /m ³
C ₁	Empirical constant for the liquid holdup	-
D	Tower diameter	m
d _{eq}	Equivalent packing diameter = 4ε/a	m
d _{nom}	Nominal packing diameter	m
d _r	Packing diameter = 6 · $\frac{\text{volume of a packing piece}}{\text{area of a packing piece}}$	m
d _{hW}	Hydraulic diameter including wall effects in small towers	m
F	Packing factor in generalized pressure drop correlation	m ⁻¹
F ₀	Packing factor at $\frac{L}{G} \sqrt{\frac{\rho_G}{\rho_L}} = 1$	m ⁻¹
F _f	Flooding factor	m ⁻¹
F _L	Packing factor at a liquid load L	m ⁻¹
f	Friction factor	
G	Gas flow rate	kg/m ² · s
g	Acceleration due to gravity	m/s ²
H	Height of packed bed	m
h	Liquid holdup	-
K _{lam}	Laminar contribution to single phase friction factor	-
K _{turb}	Turbulent contribution to single phase friction factor	-
L	Liquid flow rate	kg/m ² · s
P	Packing factor in flooding correlation of Eduljee	-
Δp	Single phase pressure drop	Pa
Δp _{Dry}	Dry pressure drop	Pa
Δp _{Wet}	Wet pressure drop	Pa
Re	Reynold's number	-
s	Standard deviation	-

v	True gas velocity	$ m/s $
v_o	Superficial gas velocity	$ m/s $
x	Constriction factor	

Greek symbols

α	Exponent for dry friction factor	
β	Constriction factor	
ϵ	Void fraction	$ - $
δ	Liquid film thickness	$ m $
δ_N	Liquid film thickness at contact points	$ m $
Λ, λ	Single phase friction factor	$ - $
μ	Dynamic viscosity	$ kg/m \cdot s $
ν	Kinematic viscosity	$ m^2/s $
ρ	Density	$ kg/m^3 $
ρ_w	Density of water	$ kg/m^3 $
ρ_{Air}	Density of air	$ kg/m^3 $
τ	Tortuosity factor	$ - $
ϕ	Correction factor for single phase flow in packings	$ - $
ζ	Ratio of Wet and Dry pressure drop	$ - $

Subscripts

G	Gas phase quantities
L	Liquid phase quantities

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AMMONIA RECOVERY FROM WASTEWATERS
IN PACKED COLUMNS

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Abstract

Wastewaters containing organic nitrogen compounds can successfully be treated with the ANAMET-process. Ammonia is formed through anaerobic fermentation and is then stripped off in a packed tower with circulating air. Desorbed ammonia is absorbed in phosphoric acid in a second tower, producing a valuable fertilizer. The ammonia desorption-absorption was studied in pilot plant experiments. At a high gas to liquid flow ratio it was possible to achieve a 95 percentage ammonia removal with a packing height of 2.7 m. Design data were established for three types of packing, Plasdek, Tellerettes, and Sulzer BX-packing. The Sulzer packing had the superior transfer efficiency, while the Plasdek had the most suitable pressure drop. The best conditions for absorption were at low pH and high salt-concentration.

Introduction

Many industrial wastewaters contain different organic nitrogen compounds, such as proteins. Wastes from baker's yeast factories may serve as an example. In these wastes the nitrogen content can be as high as 2000-3000 g N/m³. Since nitrogen promotes eutrophication of the recipients and is toxic to marine life even in low concentrations there are legal limitations to the effluent. In high concentrations it can also be worthwhile to recover and recycle the nitrogen.

Wastewaters containing nitrogen compounds can be handled with the ANAMET process¹. This process is a combined anaerobic-aerobic fermentation process which decomposes organic wastes to methane, ammonia, carbon dioxide and water. In figure 1 the procedure is schematically outlined.

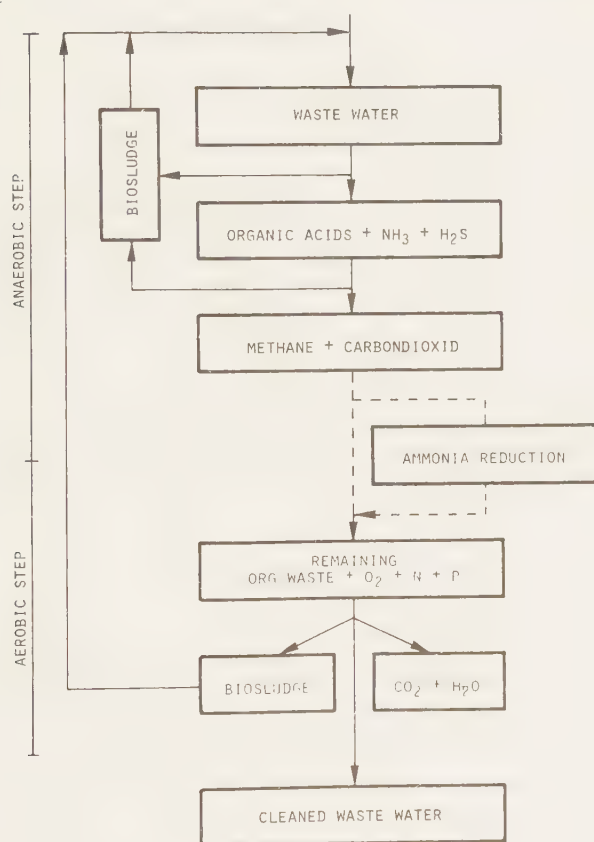


Figure 1. The ANAMET process

The ammonia is removed from the wastewater between the anaerobic and aerobic step. The removal is for instance accomplished by countercurrent desorption with air in a packed column.

Figure 2 shows more in detail the particular treatment of the wastewater when desorbing ammonia.

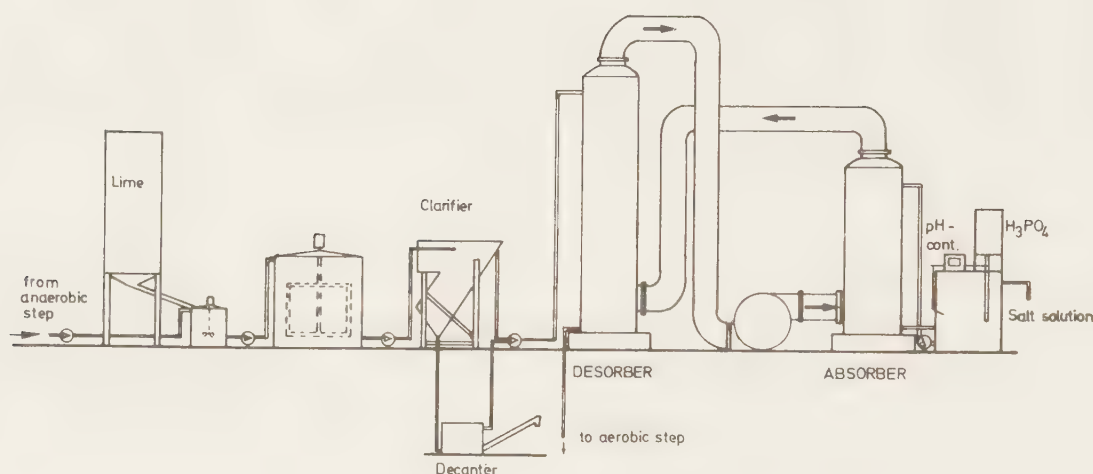


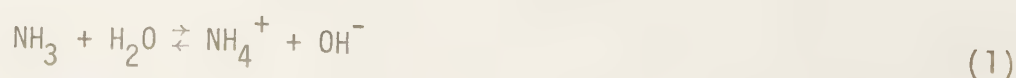
Figure 2. Layout of an Ammonia recovery system

The first step involves lime addition. The suspension is clarified in a lamella clarifier. The sludge is then dewatered and the residual liquid pumped together with the overflow from the clarifier to the top of the desorption tower, where ammonia is desorbed by air. The air is circulated and the ammonia is absorbed in a second tower. The absorption-liquid is recirculated to give the desired ammonium-salt concentration. The resulting salt may be sold on the open market or reused in the main process.

The desorption-absorption process has been extensively tested by experiments in pilot-plant scale. The results from these tests are presented in this paper. Together with theoretical considerations, essential relationships for the effective removal of nitrogen in combination with the ANAMET process are established.

General lay-out of the desorption process

In water, ammonia dissociates to ammonium-ion according to the following equilibrium reaction:



Only undissociated ammonia is possible to strip off. The quantity which is undissociated depends on pH and temperature

$$[\text{NH}_3] = \frac{[\text{NH}_3]_{\text{tot}}}{1 + \frac{10^{-\text{pH}}}{K_a}} \quad (2)$$

where the equilibrium constant, K_a , of equation (1), depends on temperature. Furthermore the solubility of ammonia in water decreases with increasing temperature. Thus the desorption is favoured by increasing pH and temperature.

In the anaerobic step of the ANAMET process the temperature and pH are kept at optimum for the conversion of wastes into methane gas. This means that practically all the nitrogen compounds exist in the ammonium-ion form. Before it can be stripped off the pH must be elevated. This is accomplished, for instance, by lime addition. To what extent the pH should be elevated is a question of economical considerations; higher pH:s require more lime but will minimize the tower height and vice versa. The temperature of the water coming from the anaerobic step is favourable but can of course be increased further by heating.

The enthalpy of the stripping air is also of importance because of the cooling effect. Existing ammonia-desorption plants consist of one tower using fresh air for the stripping. They are therefore very sensitive to climatic factors; during winter conditions the desorption efficiency is drastically reduced. Another problem is scaling occurring when carbon dioxide absorbed from the air reacts with calcium present in the water. These problems call for recirculation of the air. This is accomplished by the use of two towers; one for desorption and one for absorption. The desorbed ammonia from the wastewater is absorbed in a suitable acid in the second tower. Both towers are operated at the same temperature, eliminating cooling. The circulating air has a very low carbon dioxide content and no scaling will occur.

Ammonia is highly soluble in water. The desorption unit therefore must work with a high gas to liquid flow ratio, which makes the choice between different types of absorption-desorption units restricted. The most suitable equipment seems to be a packed tower operated counter-

currently. In such an equipment a good efficiency is combined with the highest possible driving force at a relatively low pressure drop. The type of packing must permit very high gas load and exhibit good wetting properties even at low liquid flow rates. Packings of film-type like Plasdek (AB Carl Munters, Sweden), Sulzer packing (Gbr. Sulzer AG, Switzerland) and Flexipack (Koch Eng. Co. Inc.) are very suitable, although packings of ring or saddle type of larger sizes may very well be used. In this work the Plasdek packing, the Sulzer packing type BX and the Tellerettes (Ceilcote) have been tested to give characteristic mass transfer data. A view of the packings is found in figure 3 and some geometrical data are gathered in table 1.

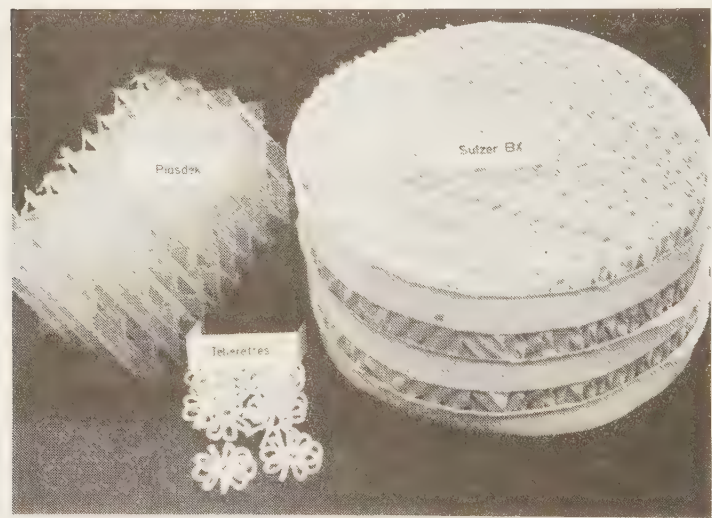


Figure 3. Tested packings

Table 1.

Characteristic data for the three packings

Packing	α	
	$\frac{2}{\pi}$	$\frac{3}{\pi}$
Plasdek 12060	12	4
Sulzer BX	4	4
Tellerettes	180	87

Theory of evaluating mass transfer data

The equilibrium partial pressure over an ammonia solution generally does not obey Henry's law. However if the concentration is less than a few percent, as in most wastewaters, the equilibrium relationship may be approximated by Henry's law. The Henry's law constant, H_e , is temperature dependent and could be expressed as²

$$H_e = 37.25 \cdot e^{-0.0525 \cdot t} \quad (3)$$

where t is the temperature in $^{\circ}\text{C}$. Since all streams were lean it is also permissible to assume that the mean driving force in the tower is equal to the logarithmic mean overall concentration difference. This means that the mass transfer coefficient, $K_G a$, for the desorption runs can be evaluated from the following equation

$$K_G a = \frac{G}{M_G \cdot z \cdot P} \cdot \frac{y_1 - y_2}{(x_1/H_e \cdot P - y_1) - (x_2/H_e \cdot P - y_2)} \cdot \ln \frac{x_1/H_e \cdot P - y_1}{x_2/H_e \cdot P - y_2} \quad (4)$$

where index 1 and 2 refer to conditions at top and bottom of the tower, respectively; x and y are the mole-fraction NH_3 in liquid and gas, respectively; P is the pressure, G the gas load, z the tower height and M_G the mole weight of the gaseous phase. The $K_G a$ -value evaluated from this equation depends on the pH at which the desorption took place. However as long as the pH is above 11 all ammonia is in molecular form and the equilibrium reaction (1) has no influence on the process. The same $K_G a$ -values can also be used together with the proper concentrations when pH is below and varying along the tower. This implies that the mass transfer properties of the liquid are independent of pH; a reasonable assumption as long as the main resistance to the mass transfer in the $\text{NH}_3\text{-H}_2\text{O}$ system is in the gas film.

The mass transfer data for most of the absorption runs were obtained from equation (5)

$$K_G a = \frac{G}{M_G \cdot z \cdot P} \cdot \ln \frac{y_2}{y_1} \quad (5)$$

This is basically the same equation as equation (4). Equation (5) implies however that the equilibrium gas phase concentration, y^* , equals zero everywhere in the column. y^* is calculated from Henry's law

$$y^* = \frac{x}{H \cdot P} \quad (6)$$

where x is the mole-fraction of molecular NH_3 . The molecular NH_3 content depends on pH and total amount of ammonia (reaction 1) in the absorbing solution. At a pH of 4 and a total concentration of 20 percent by weight of ammonium phosphate, y^* is only 0.2 ppm and could be neglected. However when pH is larger the equilibrium gas phase concentration must be considered and instead of equation (5) an equation like (4) must be used.

The absorption of ammonia in water solutions is generally both gas- and liquid-film controlled, with the main resistance in the gas-film. Generally this can be expressed with the individual mass transfer coefficients, $k_G a$ and $k_L a$ and an enhancement factor, E , as

$$\frac{1}{K_G a} = \frac{1}{k_G a} + \frac{1}{H \cdot E \cdot k_L a \cdot \rho_m} \quad (7)$$

where ρ_m is the mole-density, making allowance for the use of mole-fraction as driving-force. The relative liquid-film resistance when absorbing in pure water may be as high as 60 % of the total resistance³. When lowering the pH the relative liquid film resistance decreases because of the instantaneous equilibrium reaction. (Analogous of (1)). Eventually at sufficiently low pH the reaction proceeds at the gas-liquid interface. The process is then entirely gas-film controlled. Results of this kind are for instance presented by Feller on ammonia absorption in sulphuric acid solutions⁴.

The recirculation of the absorbing acid solution will also influence the mass transfer coefficients. According to Danckwerts⁵ and Onda⁶ an increase in ionic strength of the solution will decrease the solubility. Likewise the rate of reaction and equilibrium will be influenced. It is also possible that the ionic strength influences the mass transfer area due to changes in viscosity and surface tension of the solution⁷. No theoretical model for prediction of the total effect of ionic strength is available today. It must be evaluated by experiments.

Experimental Equipment

The experimental equipment, as displayed in figure 4 consisted mainly of

two towers; one was operated as a desorption tower and one as an absorption tower. Each of the towers had an inside diameter of 0.3 meter and could have a variable height of packing up to 3.5 meters. From a supply vessel the wastewater was pumped to the top of the desorption tower. Before entering the top the water was heated to 35°C. At the top of the tower the liquid was carefully distributed by a specially designed distributor. The NH_3 -rich air from the desorption tower was blown to the bottom of the absorption tower, where ammonia was absorbed in phosphoric acid. The ammonia content was reduced by about 90 percent before the air left the tower, and was circulated back to the desorption tower. The velocity of the circulating air was metered separately at the entrance of each tower. The phosphoric acid solution was recirculated at a pre-determined salt-concentration. The pH was controlled by adding fresh acid, accomplished by a standard pH-control system.

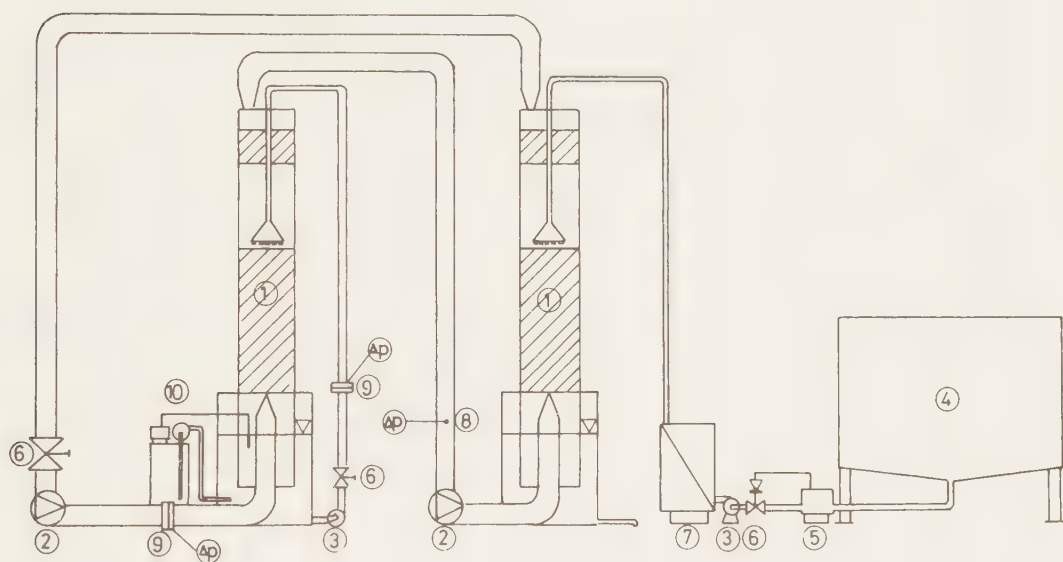


Figure 4. Experimental set-up

- | | |
|----------------------|-------------------------------|
| 1 Packed section | 6 Flow control valves |
| 2 Fans | 7 Plate type heat exchanger |
| 3 Pumps | 8 Pitot tube |
| 4 Water supply | 9 Orifice flowmeter |
| 5 Magnetic flowmeter | 10 Acid supply and pH-control |

Sampling of liquid and gas could be made at the entrance and exit of each tower. The gas samples were analyzed on-line by an IR-spectrophotometer. This was carefully calibrated down to ppm-level. The liquid samples were analyzed by an ion-selective electrode for the ammonia content. The electrode was calibrated each test day against solutions with known ammonia content. The pH of the liquid samples was also measured. Before any measurements were performed the whole desorptions-absorption system was let to thermal equilibrium, i.e. both wastewater and phosphoric acid-solution attained 35°C with the air circulating at 100 percent relative humidity.

Experiments

Wastewater from the anaerobic step of the ANAMET-process in a sugar factory was used in a first set of experiments. Since this particular wastewater only contained small amounts of nitrogen compounds, ammonia was added. The pH of the water was then elevated by lime addition, and the suspension was clarified. Although this water was artificial in respect of ammonia it contained most species and particles of various sizes normally present in real wastewaters. After desorption the ammonia was absorbed in a phosphoric acid solution at a relatively high pH. The ammonia salt can for instance be used as fertilizer. The whole test system with variables such as water, pH, temperature and packing height, thus resembled a commercial desorption-absorption system; a system that is capable of reducing the ammonia content of the wastewater to a desired level and to give a marketable product.

Since it was very impractical to use this "real" wastewater in longer test series a second set of experiments were performed with a "model" water. This "model" water consisted of tap water, ammonia and potassium hydroxide. Other test variables were held constant. A comparison of the results shows that the type of water only had a minor influence on the mass transfer properties.

However design data of mass transfer coefficient type as obtained from these two sets of experiments were very uncertain. This is because the removal of ammonia was driven too far and the gas and the liquid streams thereby approached equilibrium. By using a reduced height of the packed

section in a third set of experiments, the degree of removal was more suited to establish accurate design data. These experiments were performed with "model" water. The absorption took place at a lower pH, thus approaching total gas-film control. Finally a more comprehensive study of important absorption variables, such as pH and ionic strength, was performed. In these experiments an air-ammonia mixture was artificially made up to the same concentration as was established when the air was circulating. The absorption took place at different pH-values and salt-concentrations. A brief summary of important test parameters of the four experimental series is gathered in table 2.

Table 2. Important test parameters

		Test of "real" wastewater	Test of "model" wastewater	Tests for design data	Absorption study
DESORPTION	Packings tested	Plasdek	Plasdek	Plasdek Sulzer BX Tellerettes	
	z meter	2.7	2.7	1.50	
	pH _{in}	10.8	11.2	12	
	t °C	35	35	35	
	G kg/m ² ·s	3.3-3.5	2.9-3.5	1.7-3.4 1.7	Plasdek Sulzer Tellerettes
	L kg/m ² ·s	0.7-5	0.7-7	0.7-4	
	C _{NH₃in} kg/m ³	2.0	2.0	2.0	
ABSORPTION	Packings tested	Plasdek	Plasdek	Plasdek	Plasdek
	z meter	2.40	2.40	0.90	0.75
	pH average	6.5	6.5	2	1.5-11
	t °C	35	35	35	18
	G kg/m ² ·s	3.3-3.5	2.9-3.5	1.7-3.4	1.7
	L kg/m ² ·s	5-15	5-15	7-20	7
	Y _{NH₃in} ppm	2000	2000	2000	2000
	Salt-conc. wt-%	0-24	0-24	24	0-30

Pilot plant scale results

The experiments with "real" wastewater resulted mainly in reduction curves as displayed in figure 5.

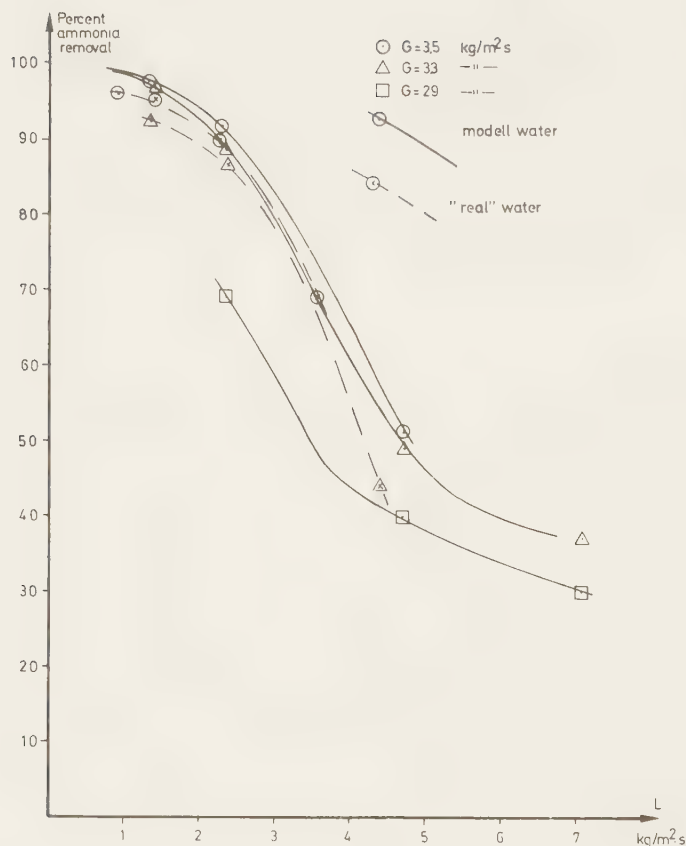


Fig. 5 Percent ammonia removal - pilot plant tests

As is apparent from the figure a good ammonia reduction was possible to establish. A high gas velocity and a low liquid load were however necessary. As previously has been mentioned the reason for this is a diminishing driving force when the gas phase tends to be saturated. One way of compensating a small driving force is of course by increasing the packing depth. However it seems to be more promising to admit liquid loads that are considerably lower than the minimum wetting rate⁸. When, simultaneously, using a gas velocity higher than 3 m/s the driving force will be satisfactorily high within the entire packed section. With the experimental set up used in this investigation it was thus possible to achieve a 95 percentage ammonia removal with a packing height of not more than 2.70 meters.

From experiments using the deeper packing bed it was also possible to conclude whether there exist any important differences in mass transfer

properties between the "real" and the "model" wastewater or not. From a theoretical viewpoint it was believed that, for instance, different surface tension and desorption of other gases than ammonia only would have a minor influence. By visual observations, during the experiments, no differences in the hydrodynamic appearance were noticed. For instance no foaming was observed when using any of the watertypes. A more comprehensive comparison may be performed by using figure 5. When looking at the degree of removal it is apparent that the ammonia more readily desorbed from the "model" water. Most of this is probably due to different starting pH-values in the two water types. Generally the "model" water had a pH greater than 11 at the entrance of the column, while the pH of the "real" wastewater was somewhat lower, which resulted in a considerably lower pH at the exit. Consequently the driving force was lower, when desorbing ammonia from the "real" wastewater, especially at low liquid flow rates where the pH-change was larger. This could be accounted for by calculating the undissociated ammonia content at either end section of the tower. By using these concentrations rather than the total ammonia concentrations when calculating the mass transfer coefficient, comparable and pH-independent k_{Ga} -values were established. Thus calculated K_{Ga} -values are found in table 3.

Table 3.

Comparison of K_{Ga} -values from "real" and "model" waste water.

G $\text{kg/m}^2 \cdot \text{s}$	L $\text{kg/m}^2 \cdot \text{s}$	K_{Ga} "real" $\text{kmol/s} \cdot \text{m}^3 \cdot \text{MPa}$	K_{Ga} "model" $\text{kmol/s} \cdot \text{m}^3 \cdot \text{MPa}$
3.3	1.4	0.99	1.10
3.3	2.4	1.24	1.17
3.5	2.3	1.25	1.30

It is obvious from the table that the mass transfer properties of the two types of water are similar. Of great importance is however to keep the pH on a high level, i.e. all of the ammonia should be undissociated.

After desorption the ammonia was absorbed in phosphoric acid solution at a pH of about 6.5. The concentration of the ammonium salt was continuously risen until the liquid was about 25 % by weight. A solution of ammonia and phosphoric acid at a neutral pH is believed to be valuable

as fertilizer. Preferably the concentration should be further increased, which is also theoretically possible. The concentration is however limited to about 38 %, where the vapor pressure reduction is of importance, i.e. water is absorbed from the about one hundred percent humidity atmosphere in the recycling gas, increasing the volym of the absorbing stream, with the salt concentration essential constant. At the temperature and pH used here this limit is lower than the concentration where the salt is precipitated. The absorption took place in a rather large quantity of liquid in order to keep the pH constant through the tower. Thus the liquid load varied from 7 to 14 kg/m²·s. The gas velocity was the same as in the desorption tower.

Finally it may be concluded from the pilot plant experiments, that the described method of recovering ammonia from wastewaters seems very promising. No problems like scaling, foaming, precipitation of ammonium salt were observed during the test period. The test period was however relatively short (approx 300 hr) and the process was run intermittently with an artificially made water. Experiments during a longer test period and under more realistic conditions will even better establish all the features of the process.

Design data

Design data were established by calculating the mass transfer coefficient for each run. However, as earlier mentioned mass transfer coefficients calculated from experiments using the deeper packing section were very uncertain. In order to establish accurate design data it was necessary to perform experiments on a smaller packing height. From a practical viewpoint, "model" wastewater was used. The entrance pH of the water was kept at about 12. The "model" wastewater then exhibits the same mass transfer properties as "real" wastewater, as earlier indicated. In figure 6 the resulting $K_G a$ -values for the desorption of ammonia are plotted as a function of liquid load with gas load as parameter.

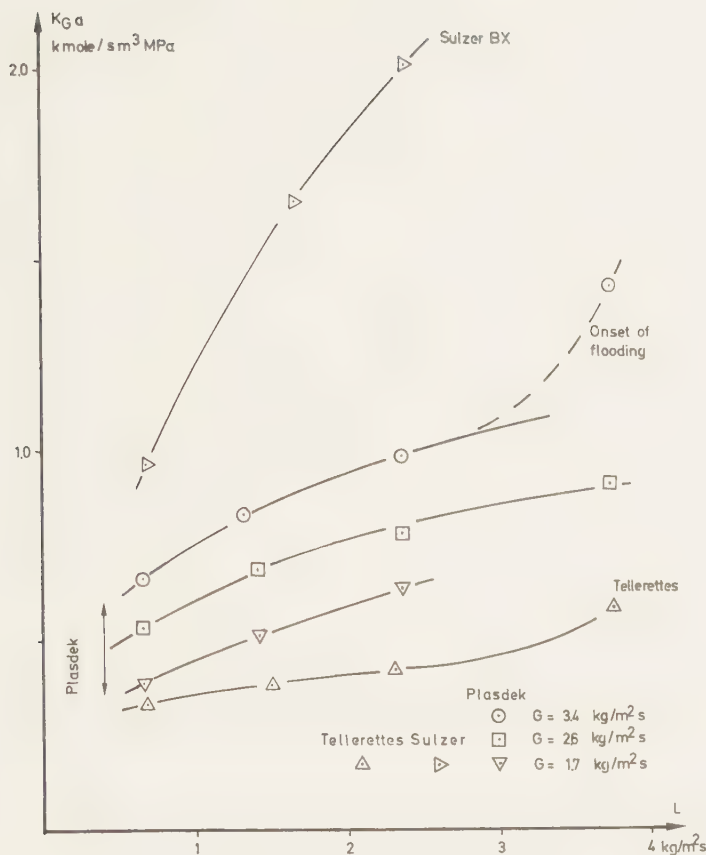


Figure 6. Design data - desorption of NH_3 from water

The mass transfer coefficients, as presented in the figure, for the three packings, are quite different. The Sulzer packing shows a considerably higher mass transfer efficiency than the others. This could partly be attributed to a twice as large specific geometric area. The special woven structure of the packing also greatly increases the efficiency; capillary forces distribute the liquid over the packing surface. At low wetting rates this is particularly important. However the ammonia containing liquid reduces the surface tension and thus partly eliminates the effect of poor wetting. The Plasdek data also show surprisingly low effect of the low liquid flow rates. The Tellerettes on the other hand seem to be more sensitive to wetting. This could probably be explained by the different way of operation. When liquid is distributed in a bed of Tellerettes droplets are formed while in Plasdek the liquid distributes in a film. At lower liquid loads the number of droplets formed may radically decrease, which then, of course, diminishes the mass transfer area.

The measurements on the Tellerettes and the Sulzer packings were only performed at the lowest gas flow rate. This limitation is due to that these packings have a low flooding point; it was impossible to use higher gas load without flooding. The Plasdek on the other hand, which has very favourable pressure drop data, allowed for higher gas loads. At the highest liquid and gas load the conditions were however very near to flooding which is evident from pressure drop measurements. The increase in mass transfer efficiency is thus probably due to incipient flooding. The tower may very well be operated at these conditions, giving better mass transfer at the expense of a higher pressure drop. When this particular experiment is omitted the ten remaining measurements with the Plasdek packing can be correlated as:

$$K_G a = 0.313 \cdot G^{0.725} \cdot L^{0.318} \quad (8)$$

The average deviation when predicting $K_G a$ using this formula is about 3 % with a maximum of 5 percent.

The experiments show that it is quite possible to use either of the three packings for the ammonia removal. The Tellerettes and the Sulzer packing however have too low void fraction and should preferably be used in a larger nominal size. This would allow for a higher gas velocity. The mass transfer coefficient of a larger packing will be somewhat lower thus requiring a higher packed section. It is also important to remember that when selecting the packing, not only the mass transfer efficiency should be considered, but also important variables like pressure drop and price.

The mass transfer coefficients obtained from the absorption measurements are presented in figure 7. Only Plasdek was investigated, since this packing permits high gas and liquid loads. A comparison between literature data and the present data shows a higher gas rate dependency and a lower liquid rate dependency. In all the experiments the absorption liquid flow rate was considerably above the minimum wetting rate. The low increase of the transfer coefficient with increasing liquid flow rate indicates that at sufficiently high liquid load the available surface of the packing is almost completely used for mass transfer. Further increase of the liquid load merely means a thicker liquid film on the packing.

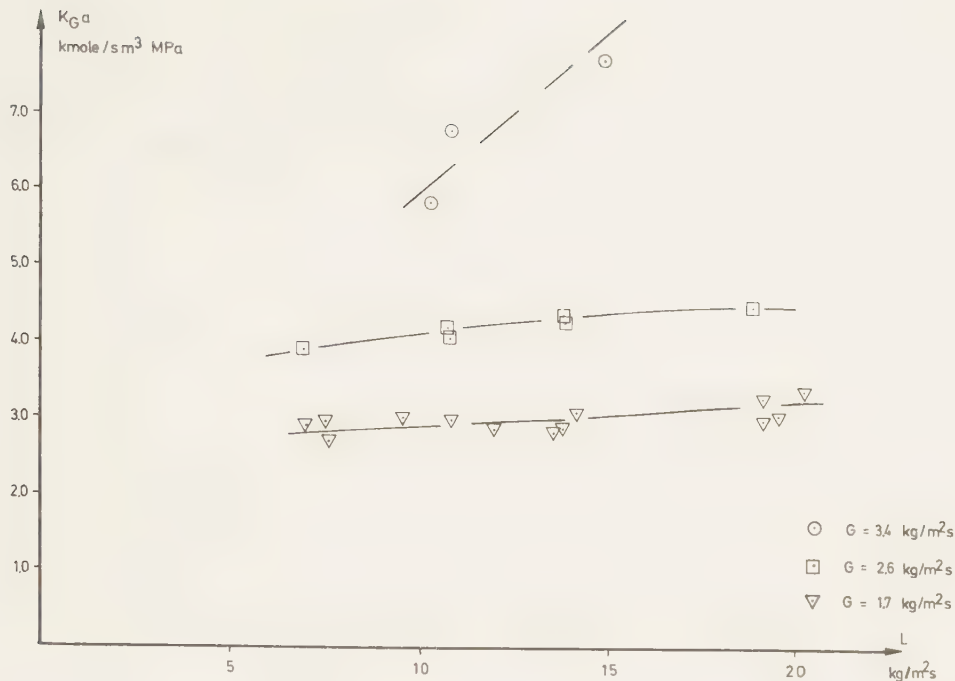


Figure 7. Design data - absorption of NH_3 in H_3PO_4 -solution

The gas flow rate was the same as in the desorption tower. The absorption tower was thus operated under conditions from loading to flooding. The K_{Ga} -values obtained at $3.4 \text{ kg/m}^2\text{s}$ clearly indicate that the packing is operated close to flooding. Also $2.6 \text{ kg/m}^2\text{s}$ is a gas velocity not normally encountered. Here the tower is operated between the loading and flooding point. Because of the higher turbulence created between the phases this means higher mass transfer coefficients than predicted from literature correlations.

The absorption of ammonia took place in phosphoric acid at a low pH. This pH is almost low enough for the ionization of NH_3 to take place at the interface between gas and liquid, i.e. the condition for full gas-film control. Sherwood and Holloway⁹ and Manford Doble¹⁰ present data on absorption of ammonia in sulphuric acid which support this anticipation. Data of this kind are however very scarce in literature. It is much more common to absorb ammonia in water^{3,4,10,11}. When absorbing in water the liquid phase resistance may be considerable. In connection with the ANAMET-process it is of interest to produce an ammoniumsalt-solution of high pH. The effect of pH on the mass transfer coefficient was separately studied by absorption of ammonia in three types of liquid; in tap water without any buffer action, in water with different amounts of phosphoric acid and high salt-concentration and in water at a higher

constant pH. The pH-effect was studied using one liquid to gas load ratio, selected to minimize the pH-change through the tower. In spite of this the pH could vary up to 0.5 units. When absorbing in water pH varied between 7 and 10. In figure 8 the experimental results are summarized.

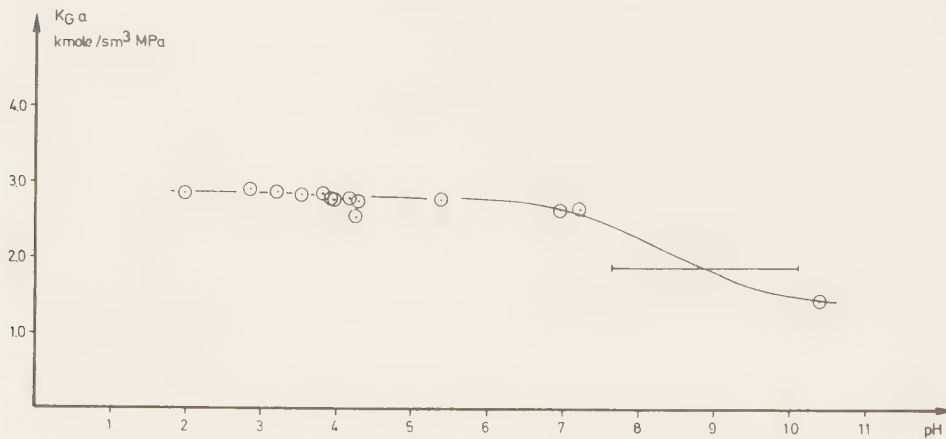


Figure 8. Influence of pH (salt-conc. > 25 wt-%)

From this figure the decrease in mass transfer coefficient with increasing pH directly may be established. In the pH-range of primary concern for the ANAMET application (2-7) the mass transfer coefficient is only altered about 5 percent. As long as the absorbing liquid contains protons at the outlet from the tower, all ammonia absorbed is ionized somewhere in the liquid film resulting in a K_{Ga} -value only slightly different from the gas-film coefficient. However when the absorbent is ordinary water the protons are soon consumed, which means that the ammonia molecules must diffuse through the entire liquid film. This results in a larger liquid-film resistance and a lower K_{Ga} -value. Likewise at higher pH the solubility decreases because the ionization is suppressed giving even more liquid-film resistance. The relationship between pH and K_{Ga} presented in figure 8 is of course a function of gas to liquid flow ratio. In the pH-range and salt-concentration applicable for the ANAMET process, however, smaller changes in the flow ratio are believed to have only minor influences.

As earlier mentioned the ionic strength of the absorbing solution is also of importance. By continuously adding fresh phosphoric acid to the absorbent at a constant pH and liquid to gas flow ratio, this effect was also studied. Figure 9 gives the resulting relationship for two different pH:s.

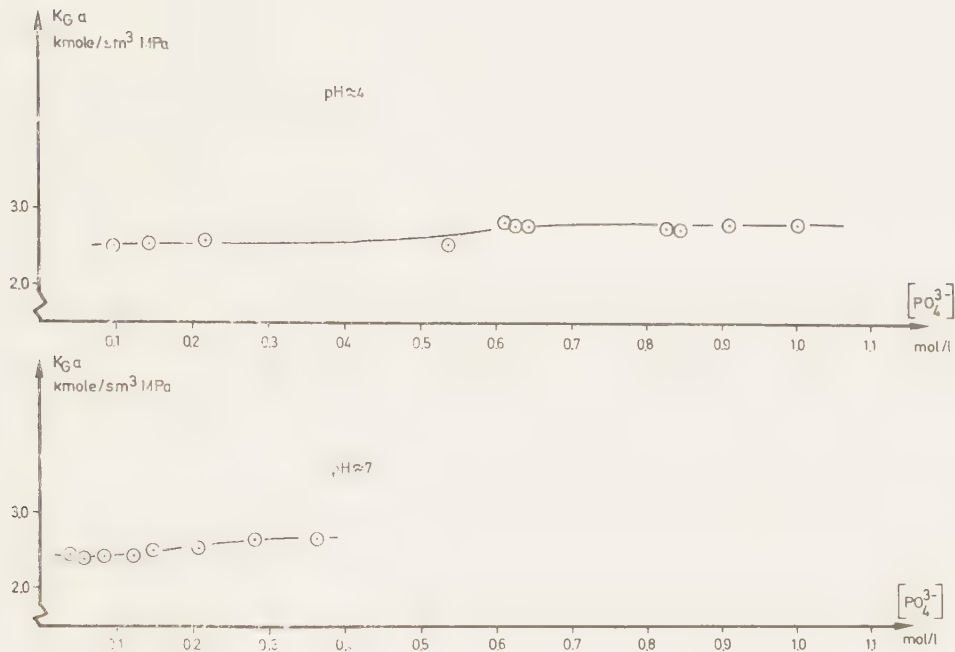


Figure 9. Influence of salt-concentration

The same type of relationship was obtained; a small increase of mass transfer efficiency until a stable value is reached. The important conclusion is that the efficiency increases in spite of decrease in solubility. Changes in rate constant, diffusion coefficients, equilibrium value and interfacial areas seem to be dominant. The increase is however not very marked, only about ten percent. The stable $K_G a$ -value is also reached at a relatively low salt-concentration. At higher concentrations $K_G a$ -values presented in figure 7 may be used together with the pH-correlation in figure 8 to establish design data for the absorption of ammonia at different pH-values.

Some economic aspects of the ammonia removal system

Some important cost items connected with the ammonia removal system are gathered and displayed in figure 10¹².

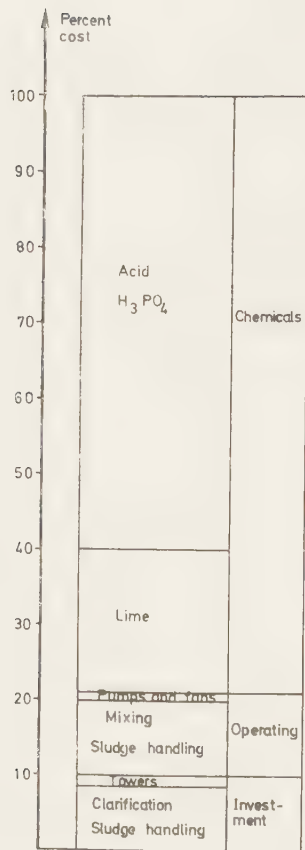


Figure 10. Various annual cost items (Plasdek)

It is obvious that the main contributors are the lime and acid costs. It is however to some extent possible to reduce the lime cost by using a lower pH when desorbing the ammonia. Likewise a sound choice of pH in the absorption tower reduces the cost of acid. The minimization of lime and acid consumption means higher towers. The operating and investment costs of the towers on the other hand only contribute to the total cost with a few percent.

Another way of reducing the acid cost is to choose a cheaper acid, like sulphuric acid. It should however be borne in mind that ammonium phosphate is a valuable fertilizer and thus may give an income to the process. The total cost per cubic meter when using sulphuric acid is well below the cost for other applicable ammonia removal systems, for instance biological denitrification.

Conclusion

The presented stripping method for ammonia reduction seems very promising. Many serious drawbacks of straight through stripping are avoided by circulating the air, and absorbing the ammonia in H_3PO_4 . The results indicate that a high gas velocity and a low liquid flow rate give the best desorption performance. Only packings of larger sizes can then be used. Even then the liquid load must preferably be lower than the minimum wetting rate. This does however not seriously affect the mass transfer because the ammonia containing solution distributes better than water. The absorption is favoured by a low pH and a high ionic strength. The economics of the process seem to be comparable to other ammonia reduction systems. Care should be taken in selecting pH because of high cost of lime and acid.

Acknowledgements

Credits

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Nomenclature

A	Concentration of A	kmole/m^3
a	Specific geometric area of packing	m^2/m^3
C	Concentration	kg/m^3
E	Enhancement factor due to reaction	--
G	Gas flow rate	$\text{kg/m}^2 \cdot \text{s}$
He	Henry's law constant	$1/\text{MPa}$
K_G^a	Overall mass transfer coefficient	$\text{kmole/s} \cdot \text{m}^3 \cdot \text{MPa}$
k_G^a	Individual gas-film coefficient	$\text{kmole/s} \cdot \text{m}^3 \cdot \text{MPa}$
k_L^a	Individual liquid film coefficient	$1/\text{s}$
K_a	Equilibrium constant	m^3/kmole
M_G	Gasous molecular weight	kg/kmole
L	Liquid flow rate	$\text{kg/m}^2 \cdot \text{s}$
P	Absolute pressure	MPa
t	Temperature	$^{\circ}\text{C}$
x	Mole-fraction NH_3 in liquid	--
y	Mole-fraction NH_3 in gas	--
z	Tower Height	m
ϵ	Void fraction of packing	--
ρ_m	Mole-density of liquid	kmole/m^3

Subscripts

1	Top section of the tower
2	Bottom section of the tower
*	Equilibrium value
tot	Total amount of NH_3

